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Impact of anthropogenic and natural processes on the degradation of water and sediment quality of the Mghohga River estuary (northern Morocco)

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Abstract: The Tangier city knew in the last decades an increasing production of household and industrial waste caused problems critical pollution. The objectives, in this study, were to understand the monitoring of water and sediment quality in Tangier area based on the comparison of hydrochemical data variations and land use changes during the urbanization, to distinguish the impact of natural processes and anthropogenic activities on the water and sediment quality. Six metallic trace elements were investigated at 5 water and sediment sites during 2010 and 2012. Concentrations of selected metals were measured using an atomic absorption spectrophotometer (AAS). The results obtained show that the level of the station where the discharged wastewater discharges from the Tangier city, there has been a significant change in the values of the principal elements (Cr and Cd). Indeed, the metal content of the sediments of the Mghogha estuary of these elements reach particularly high values at three levels: in the vicinity of discharges upstream and downstream of the Mghogha estuary respectively characterized by a strong industrial activity and a rejection of domestic wastewater. Significant correlations ($p<0.05$) between Cr-Cd were observed in sediment and Pb-Fe in water.

Key words: Tangier, sediments, Water, heavy metals

Introduction

Currently, we are witnessing an expansion and an increase of the industry and agricultural activities, as well as a rapid growth of the population. The internal discharges of the human activities (industrial and domestic) sometimes untreated are carried by water runoff to the marine environment (Achab *et al.*, 2007; El Morhit *et al.*, 2011). Rapid industrial

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development, as well as the use of metals in production processes has led to the increased discharges of heavy metals into the environment (Avenant-Oldewage and Marx 2000).

Human activities commonly affect the distribution, quantity, and chemical quality of water resources. The range in human activities that affect the interaction of groundwater and surface water is broad (Berny *et al.*, 2002). Indeed, water plays an important role in the Moroccan economy: including tap water by humans. However in the last decades, we noticed the deterioration of stream water quality due to industrialization and human activities (El Morhit *et al.*, 2013). In Morocco, the increasing production of the household and industrial waste causes critical problems of the pollution. The complex and heterogeneous nature of household waste involves difficulty for their treatment and management (Rassam *et al.*, 2012).

Many organic pollutants and various trace metals introduced into aquatic environments concentrate in living organisms at levels sometimes exceed their rates in sediments and can therefore lead to problems of environmental, health and economic (Meddtl, 2011; El Morhit *et al.*, 2011).

The heavy metals can be recycled by chemical and biological processes, between the two compartments: water and sediment. The accumulation of metal contaminants in sediments can cause serious environmental problems (Carman *et al.*, 2007). The concentrations of heavy metals in sediments are high and associated with significant anthropogenic activities (Zourarah *et al.*, 2007; Carman *et al.*, 2007; El Morhit *et al.*, 2008).

In Moroccan estuaries, several authors have in evidence put the contaminations that are due to domestic, industrial and agricultural activities (Belbachir 1997; Tahiri 2005, El Morhit *et al.*, 2013).

In this study, we conducted a study of the impact of certain domestic, industrial, agricultural and human activities on water and sediment quality of the Mghogha River estuary, and through a diagnostic the current situation of pollution and rigorous monitoring of its evolution during two consecutive years (2010 - 2012).

Materials and methods

Study site

The bay of Tangier is located in the north-western extremity of Morocco, on the southern border of the Gibraltar Strait, between parallel of 35°46' and 35°48' North and meridians of 5°45' and 5°49' West. Corresponds to a maritime depression, limited under the effect of the neotectonic by two rocky tips: the Cape Spartel on the West, and the Cape of Malabata in the East.

Tangier bay is characterized by an important hydrographic network, constituted by rivers

(continental emissaries) with relatively weak flow, crossing from south toward the north the urbanized and industrialized zones of Tangier city. These rivers by order of hydrological and hydraulic importance are Mghogha River (Fig. 1). From the estuary of this river, five stations were investigated, related to the presence of industry (congealment, cannery, and fish flour), active fishing sites, garbage of factories, agriculture and domestic worn-out waters (Fig. 2):

- Station 1: located at the main road and highway 15 km from the mouth. The catchment of this station is 35°45'11N, 005°47'24O with altitude is the 9m.

- Station 2: located 10 km from the mouth. The catchment of this station is 35°46'15N, 005°46'50O with altitude is the 5m.

- Station 3: it is distant 7 km from the mouth. The catchment of this station is 35°46'24N, 005°46'51O with altitude is the 2m.

- Station 4: located 3 km from the mouth. The catchment of this station is 35°46'30N, 005°46'53O with altitude is the 4m.

- Station 5: located at the mouth. The catchment of this station is 35°46'35N, 005°46'55O with altitude is the 3m.

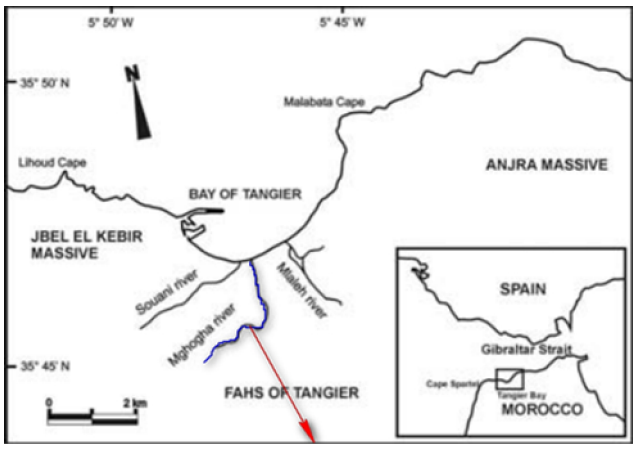

Fig. 1: Geographical situation of the study area.

Fig. 2: Site of sample collection (Mghogha River estuary in Morocco).

Sediment

Core samples up to 20 cm in length were collected from 15 sampling sites in March, May and July 2010 and 2012 (2 sampling stations in each of the 5 stations selected) using polyvinyl chloride corers (Cabrera *et al.*, 1992; El Morhit 2009). The corers were immediately sealed and stored at 4°C until arriving at the laboratory. In the laboratory, the cores were extruded and sectioned. The first 5 cm section of each core was used in this study (Meyerson *et al.*, 1981).

Sections were air-dried (Thomas *et al.*, 1994) and sieved with a 63-µm nylon mesh, and the fraction <63 µm was chosen for chemical analysis (Rauret *et al.*, 1988; Thomas *et al.*, 1994). The samples underwent acid digestion (HNO_3-$HCLO_4$) in an automatic microwave digestion system. Trace metals (e.g., copper, chromium, lead and cadmium) in digested sediment samples were determined by a Graphite Furnace atomic absorption Spectrophotometer (AAS) (AWG 120) and iron and zinc contents were analyzed by flame (VARIAN AA 240 Z).

Blanks were included in each batch of analysis. Calibration standards were regularly performed to evaluate the accuracy of the analytical method

Water

Water samples were collected from the same sampling sites as the sediment, using 1l acid-leached poly-ethylene bottles. The samples collected in March, May and July 2010 and 2012 were filtered through a 0.45 µm membrane filter, acidified with 2 ml of concentrated HNO_3 and stored at room temperature until analysis. The samples were analyzed by graphite furnace AAS, with Zeeman background corrector, after preliminary metal concentration according to the method described by Sturgeon *et al.*, (1980). Copper, lead and cadmium was analyzed by Graphite Furnace atomic absorption Spectrophotometer (GFAAS) (Perkin Elmer 4100ZL) using Zeeman-effect background correction. Iron and zinc were analyzed by Flame-AAS. Chromium was analyzed directly, without pre-concentration, by GFAAS (Perkin Elmer 4100ZL) using Zeeman-effect background correction and operation conditions recommended by the manufacturer. Sample concentrations of chromium were, however, below the detection limit (0.42 µg/l) and are not presented here.

Reagents and quality assurance

All reagents used were of analytical grade (Merck). Standard working solutions of the different elements analyzed were prepared from the corresponding 1000 mg/l Merck titrisol solution.

The digestion and analytical procedures were checked by analysis of standard reference materials (sediment: CRM-277, Community Bureau of Reference; and seawater: CASS-3, National research Council Canada). Replicate analysis of these reference materials showed good accuracy, with recovery rates for metals between 92% and 98% for sediment and between 95% and 102% for water.

Statistical analysis

In the present study, both a Person Correlation was conducted using XLSTAT for windows release 12.0.

Results

Sediment

The highest concentration levels of most of the analyzed metals (iron, zinc, lead, chromium, copper and cadmium) were found in the sediment from the station 1, due to fact that this station was polluted by toxic industrial wastes (Tab. 1). Higher mean iron concentrations (3593.30 µg/g dry mass) were obtained in the station 1, and lower mean cadmium concentrations (2.26 µg/g dry mass) were detected in station 5. The sediment metal concentration followed the order: (S5) > (S1) > (S3) > (S4) > (S2). The general ranking of metal accumulation in sediment was: Fe > Zn > Pb > Cu > Cr > Cd (Fig. 3). The highly significant positive correlation was obtained between Cr-Cd in sediment ($p \leq 0.05$; Tab. 2).

Tab. 1: Mean concentrations (µg/g dry mass) of heavy metals in sediment.

Metal	S1	S2	S3	S4	S5
Zn	87.23±40.90	73.79±51.54	26.60±10.02	26.50±7.09	297.72±279.72
Pb	73.49±61.51	63.28±60.49	29.27±27.09	19.49±17.00	84.27±81.94
Fe	3593.30±3589.75	3225.02±3221.65	3569.18±3567.77	3396.42±3395.39	3482.37±3482.01
Cr	16.77±1.35	3.92±1.57	2.24±1.88	4.11±0.70	66.15±61.68
Cu	31.01±19.23	26.02±19.97	10.88±5.828	8.31±2.76	50.57±45.26
Cd	2.80±1.47	2.73±1.49	2.85±1.76	2.88±1.71	2.26±2.13

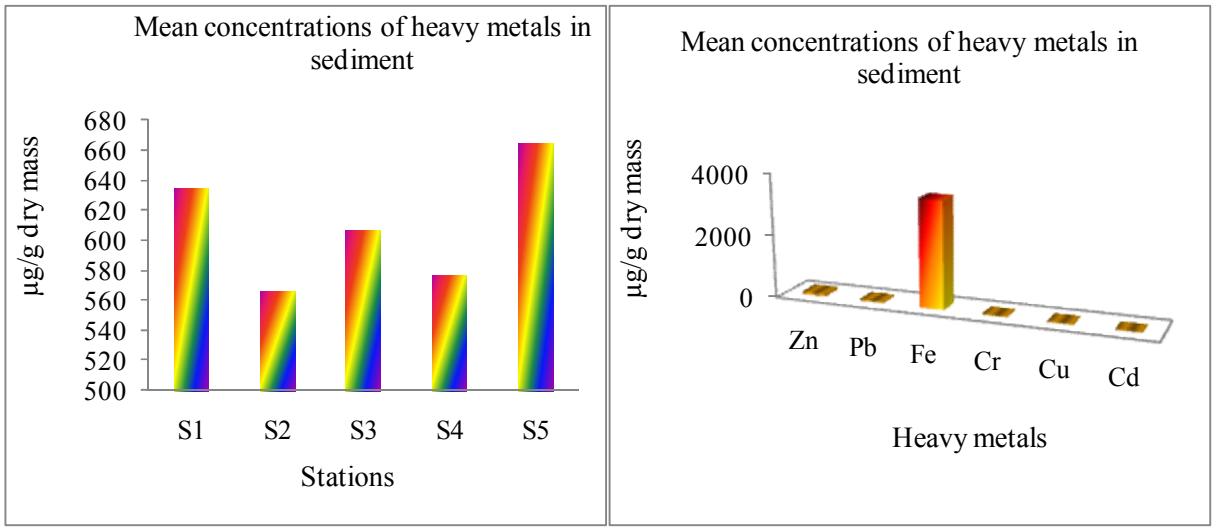

Fig. 3: The metal concentration of heavy metals in sediment.

Tab. 2: Pearson Correlation matrix among monitored metals in sediment. The values with star (*) are significantly different from 0 (level of significance alpha = 0.05).

	Zn	Pb	Fe	Cr	Cu	Cd
Zn	1					
Pb	0.78*	1				
Fe	-0.47	-0.81*	1			
Cr	0.97*	0.62	-0.30	1		
Cu	0.93*	0.94*	-0.65*	0.84*	1	
Cd	0.97*	0.67*	-0.42	0.98*	0.86*	1

Water

Metals concentrations in water are given in Table 3. Higher concentrations of lead in water were detected in station 1 (0.21 µg/l). Bearing in mind the absence of industrial dumping of lead in the area, this may be due to a heavy-traffic road across the station. Concentration of chromium, copper, lead, cadmium and iron were significantly higher in the water from the station 1 than in station 5. Higher mean chromium concentrations (70 µg/l) were obtained in station 1. However, lower mean zinc lead (20 µg/l) were noted in several stations. The water metal concentration followed the order: (S1) > (S5) > (S2) > (S4) > (S3). The general order of monitored metal accumulation was: Pb > Cu > Cd > Cr > Fe > Zn (Fig. 4). Significant positive correlation was found between Pb-Fe in water ($p \leq 0.05$; Tab. 4).

Tab. 3: Mean concentrations (µg/l) of heavy metals in water from five sampling stations.

	S1	S2	S3	S4	S5
Zn	30	20	20	20	20
Pb	210	19	24	150	120
Fe	40	40	45	20	10
Cr	70	67	65	40	52
Cu	190	60	15	25	190
Cd	10	170	79	22	30

Tab. 4: Pearson Correlation matrix among monitored metals in water.

	Zn	Pb	Fe	Cr	Cu	Cd
Zn	1					
Pb	0.14	1				
Fe	0.39	0.89*	1			
Cr	0.48	0.66	0.80	1		
Cu	-0.16	-0.34	-0.45	-0.86	1	
Cd	0.09	-0.35	-0.53	-0.72	0.84	1

The values with star (*) are significantly different from 0 (level of significance alpha = 0.05).

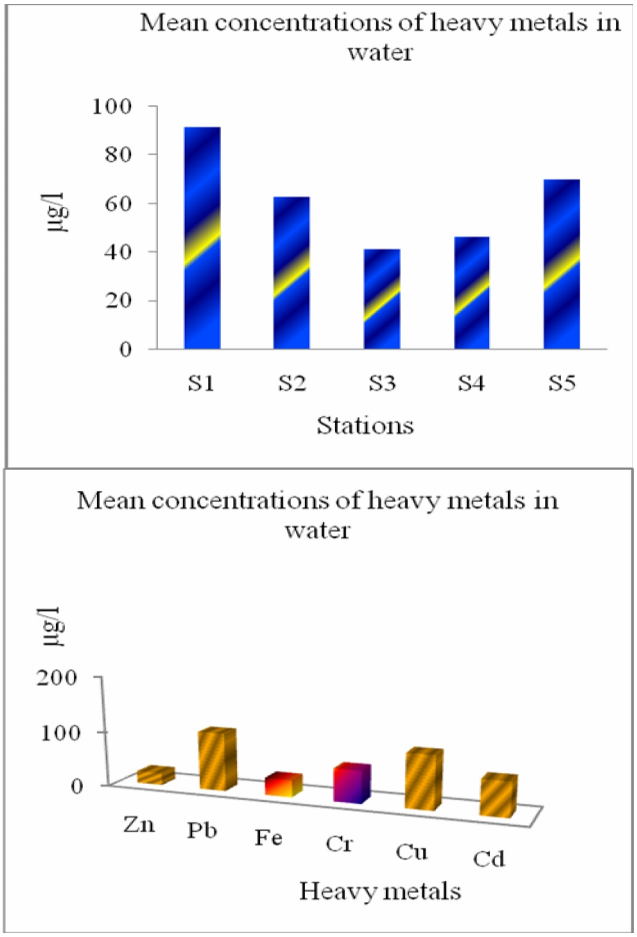

Fig. 4: The metal concentration of heavy metals in water.

Discussion

Sediment

High concentrations of iron, zinc and lead were found in the sediment from the station 1 and were comparable only to those obtained in other areas affected by pyrite pollution, such as the Huelva estuary (Pérez *et al.*, 1991; El Morhit *et al.*, 2009), or because of significant industrial dumping as in Gdansk Bay in Poland (Glasby and Szefer, 1998). By contrast, in stations 2 and 4, the levels of these metals were low, similar to those found in sediment from other non-polluted areas (Forstner and Wittman, 1983). An exception to this was lead, which reached considerably lower concentrations (19.49 µg/g dry mass) in the station 4 than in station 4 (63.28 µg/g dry mass). This is because the station 2 is near a heavily travelled highway and probably as has been documented in another study (El Morhit *et al.*, 2008). The increase of the salinity in this station contributed to the release of lead from sediment to water (El Morhit *et al.*, 2009).

Among the metals analyzed in Mghogha River estuary, the levels of cadmium in sediment are the lowest in the five stations. Data on cadmium in Moroccan aquatic systems are less abundant than for other metals (El Morhit *et al.*, 2013). The few recent studies that were carried out in estuaries (Tahiri *et al.*, 2005) pointed out the role of human activities in cadmium loadings, in agreement with other studies in Pennsylvania (USA) (Bopp and Biggs, 1981).

The amounts of iron are high in all the studied stations, consistent with previous studies in Morocco (Texier *et al.*, 1994; Cheggour *et al.*, 2005).

The high concentrations of iron found in the sediment could have resulted in the high Fe levels in the water during the process of release. Iron is the most abundant metal in Mghogha River estuary because it is one of the most common elements in the earth's crust and the sediment from the station 5 has high pyrite

(FeS2) content (Snoussi, 1984; El Morhit, 2009). According to Cabrera *et al*. (1999), pyrite oxidation produces sulphate and the Fe^{2+} ion, which is oxidized to Fe^{3+} by microorganisms such as *Thiobacillus ferrooxidans*.

High concentrations of chromium were found in the sediment from the station 5 and were comparable only to those obtained in other areas affected by industrial wastewater pollution, such as the Om Rbiâ estuary (Jadal *et al*., 2002), Port Jackson estuary (Hatje *et al*., 2003) and Bouregreg estuary (Tahiri *et al*., 2005).

Water

In the water, the metal contents measured in stations 1, 5 and 2 were considerably higher than those previously reported in marine water (Nicolai *et al*., 1999; Cotté-Krief *et al*., 2000; El Morhit *et al.,* 2009). By contrast, the station 1, is affected by contamination from the waters of draining of the rice fields (Blinda, 2007), showed zinc, lead, iron and cadmium concentrations that agree with those reported in other studies for coastal and estuary waters. For example, the mean lead levels of 104 µg/l in stations in our study are higher to those reported by Usero *et al*. (2003) in salt marshes on the southern Atlantic coast of Spain and by El Morhit *et al*., 2009 in Loukkos River estuary. Works done by Morales *et al.* (1999) and by Usero *et al*. (2003) had shown cadmium levels similar to those in our study. Copper concentrations (67 µg/l in station 2 and 15 µg/l in station 3) were of the higher order of magnitude as those found by Nicolai *et al.* (1999) in the Rhone River estuary in France and Usero *et al*. (2003) in salt marshes on the southern Atlantic coast of Spain. The range of chromium levels in our study was 40 µg/l (in station 4) to 7 0 µg/l (in station 1). These values are higher than those recorded by El Morhit *et al.* (2009) in the Loukkos River estuary (4-7 µg/l). These values are higher than those recorded by Morales *et al.* (1999) in the Gulf of Valencia (0.16 µg/l). However, chromium, levels higher than ours were found in the Po River estuary in the Adriatic Sea (Italy) by Pettine *et al*. (1997). The chromium concentration in the water was much higher. This could be suggestive of a large quantity of chromium uptake via the bottom (Avenant-Oldewage and Marx, 2000).

Metals correlation between sediment and water

The results obtained for the water and sediment study done at upstream and downstream Mghogha River estuary, showed that the water quality at upstream (S1 and S2) was poorer than the water quality at downstream (S4 and S5), with sediment contamination higher at downstream. This result is similar with those obtained by several authors (El Morhit *et al*., 2012).

The assessment of trace metals and their correlations in Mghogha River estuary reflected the degree of pollution, which is considered by many regulatory agencies to be one of the largest risks to the aquatic environment. The mean levels of iron, zinc, copper, chromium, lead and cadmium in the Mghogha River estuary were lower than the guidelines for drinking-water quality (WHO, 2006), which the mean (Fe, Zn, Cu, Cr, Pb and Cd) concentrations in the water consumption are higher (300, 3000, 1000, 50, 10 and 3 µg/l, respectively).

Based on guidelines, direct use of water from the Mghogha River estuary without treatment may aggravate poor health of sensitive groups (Savory and Wills, 1991). For example, the criterion for lead in water for domestic use is 0 to 1.70 g/l (FEPA, 1991). At levels > 100 g/l, possible neurological damage in fetuses and young children may occur (Fatoki *et al.,* 2002). For some of the metals studied in the surface sediments, the metal levels in our study were higher than the metal levels in the sediments of Montevideo Harbour, Uruguay (Muniz, *et al*., 2004), Taylor Creek, southern Nigeria (Okafor and Opuene, 2006), Bouregreg estuary (Tahiri *et al.,* 2005) and Om Rbiaâ estuary (Jadal *et al.,* 2002).

Lower concentrations of lead (63.96 µg/g dry mass) were found in the sediment from Mghogha River estuary, compared with those recorded by RNO (1995) in France (60 mg/kg dry mass). But higher concentrations of cadmium (2.70 µg/g dry mass) were found in the sediment from Mghogha River estuary, compared with those recorded by RNO (1995) in France (0.15 mg/kg dry mass).

Furthermore, the study sites are located in the discharge points of Mghogha River estuary reflecting the cumulative effects of dispersed inputs from Mghogha River and other. In spite of the levels of trace metals in the sediments, we can deduce that the sediments presented concentrations that were at the Severe Effect Level (Persaud *et al.,* 1992) and may cause adverse biological effects except for iron, zinc and copper respectively.

Thus, the contamination changes between water and sediment have resulted in a serious degradation of the ecosystem. It is suggested that modernized irrigation technology and new regulation to cover water resources management and allocation within the river basin are urgently needed to achieve sustainable development (Ma *et al.,* 2005; El Morhit *et al.,* 2008).

Conclusion

Analytical results showed that the heavy metals contents of the water are below the maximum limits set by World Health Organization (WHO) for water. However, the limits for chromium and cadmium remain higher in sediments. And by the release phenomenon, the water will be polluted by these elements.

Through analysis of the mean of the metallic trace elements of the water in the Tangier River estuary, the current state of water quality, reflects the presence of a moderate to permanent pollution and localized near the city of Tangier with significant spatiotemporal variations.

Given the heavy load produced by industrial activity and urban highly active. Several questions take such as: Absent of a significant and substantial impact on the hydrochemistry of the water system since the concentrations measured in water do not reflect accurately the relative contributions of pollution. This could be explained in the intervention of some physico-chemical phenomena such as precipitation, trapping, settling and storage of pollutants in sediments without neglecting the phenomena of bioaccumulation in fauna and flora.

The matrix correlation results between water and sediment can be categorized by five major factors: (1) Holocene transgression and mixing; (2) surface water infiltration; (3) multi-factor processes; (4) rainfall and agricultural fertilizer contamination; and (5) Geogenic Factor enrichment. This study demonstrates that the great variation of water hydrochemistry in the Mghogha River estuary should be attributed to both natural and anthropogenic processes. Indeed, an investigation on the latter subject is under study by our research group to assess completely the situation in Mghogha River estuary.

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