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Quality of Moroccan Atlantic coastal waters: water monitoring and mussel watching

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Abstract − The quality of Moroccan Atlantic coastal wasters was monitored from 1993 to 1997 by measuring hydrological parameters (dissolved oxygen, suspended particulate matter, phosphates, and nitrates), and using the mussel *Mytilus galloprovincialis* as a quantitative bioindicator of cadmium, copper, manganese, and zinc contamination. Mean concentrations of dissolved oxygen and suspended particulate matter were indicative of the effects of urban and industrial discharges of wastewater, particularly at Mohammedia, Casablanca, Mehdiya, Jorf Lasfar and Safi. Stations receiving urban wastewater showed high nitrate concentrations, especially at Mehdiya, Rabat, Mohammedia, Casablanca and El Jadida. Metal concentrations in mussels showed significant variations depending on the station and sampling period. Jorf Lasfar and Safi had the highest mean concentrations for cadmium (8 and 7 $\mu g \cdot g^{-1}$ dry weight respectively) and copper (74 and 25 $\mu g \cdot g^{-1}$ dry weight). The relation between cadmium concentrations in mussels, and phosphate concentrations in water suggests that the processing of phosphate ores at these two sites is responsible for contamination. Mussels in the Mohammedia–Casablanca sector had the highest zinc concentrations (338–379 $\mu g \cdot g^{-1}$ dry weight), followed by those collected at Jorf Lasfar (267 $\mu g \cdot g^{-1}$ dry weight), and Sidi Moussa and Safi (290–301 $\mu g \cdot g^{-1}$ dry weight). The distribution of manganese concentrations, which were maximal in estuaries (up to 19 $\mu g \cdot g^{-1}$ dry weight), was indicative of terrigenous inputs. Seasonal variations in mussel metal concentrations were characterised by winter minima, and apparently related to the physiological cycle of the animal. With the exception of cadmium-contaminated areas, the quality of mussels on the Moroccan Atlantic coast is good with respect to food safety standards. © 2001 Ifremer/CNRS/Inra/IRD/Cemagref/Éditions scientifiques et médicales Elsevier SAS

bioindicator / monitoring / mussel watch / nutrients salts / trace elements / water quality

Résumé − Qualité physicochimique des eaux côtières et bioaccumulation métallique chez la moule *Mytilus galloprovincialis* sur le littoral atlantique marocain. La qualité des eaux côtières atlantiques marocaines a été suivie de 1993 à 1996 par la mesure des paramètres hydrologiques (oxygène dissous, matière en suspension, phosphates et nitrates) et l'utilisation de la moule (*Mytilus galloprovincialis*) comme bioindicateur quantitatif de la contamination par le cadmium, le cuivre, le manganèse et le zinc. Les teneurs moyennes en oxygène dissous et matière en suspension révèlent la trace des émissaires d'eaux usées urbaines et industrielles, notamment à Mohammedia, Casablanca, Mehdiya, Jorf Lasfar et Safi. Les stations recevant des eaux usées urbaines, accusent d'importantes teneurs en nitrates, en particulier au niveau de Mehdiya, Rabat, Mohammedia, Casablanca et El Jadida. Les teneurs métalliques chez la moule montrent des variations significatives selon les stations et la saison de prélèvement. Les sites de Jorf Lasfar et Safi, avec des concentrations moyennes respectivement de 8 et de 7 $\mu g \cdot g^{-1}$ (poids sec) en Cd, et de 74 et de 25 $\mu g \cdot g^{-1}$ (poids sec) en Cu, se distinguent des autres stations. La relation entre les concentrations en Cd dans les moules et les concentrations en phosphate dans les eaux suggère que le traitement des minerais de phosphates présent dans ces deux sites est à l'origine de cette contamination. Ce sont les moules de Mohammedia–Casablanca qui sont les plus chargées en Zn (338–379 $\mu g \cdot g^{-1}$, poids sec), puis celles collectées à Jorf Lasfar (267 $\mu g \cdot g^{-1}$, poids sec), Sidi Moussa et Safi (290–301 $\mu g \cdot g^{-1}$, poids sec). La distribution des concentrations en Mn, montre des maxima dans les estuaires (jusqu'à 19 $\mu g \cdot g^{-1}$, poids sec), et trace les apports terrigènes. Les variations saisonnières des métaux chez la moule, caractérisées par des minima hivernales, semblent être liées au cycle physiologique de l'animal. Du point de vue sanitaire, à l'exception des zones contaminées par le Cd, la qualité des moules du littoral atlantique marocain est bonne. © 2001 Ifremer/CNRS/Inra/IRD/Cemagref/Éditions scientifiques et médicales Elsevier SAS

bioindicateur / suivi / monitorage quantitatif / sel nutritif / elément-trace / qualité de l'eau

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1. INTRODUCTION

The Moroccan coast, like many other coastal regions in the world, is developing rapidly with respect to the socio-economic interests of the country. Demographic expansion, and continual urban and industrial development are exposing coastal waters to increasing contaminant discharges that convey various chemicals, including metals, into the sea.

The main purpose of this study was to evaluate the physicochemical quality of the environment and the state of metal pollution in an area of relatively intense human activity. It was performed during several annual cycles (1993 to 1997) along a coastal area running from the town of Larache on the North Atlantic seaboard, to Safi in the centre, a distance of nearly 400 km. This assessment involved hydrological studies and analysis of certain metals in tissues of the mussel *Mytilus galloprovincialis*, selected as a bioindicator of the degree and extent of chemical pollution. This work can thus be considered in part as a Moroccan ‘mussel watch’. Mussels are used worldwide as a bioindicator in monitoring programmes (e.g., Anon., 1980; Cossa, 1989; Stronkhorst, 1992; Cantillo, 1998). Moreover, various ecotoxicological studies have been concerned with mussels, which are frequently consumed by man, and play an important role in the transfer of chemical contaminants along food chains. One of the main environmental problems with toxic metals is their capacity to accumulate in the tissues of living organisms, particularly bivalve molluscs collected for human consumption. The absorption of large amounts of metal by edible species could ultimately be harmful to man at the upper end of the food chain.

2. ENVIRONMENTAL SETTINGS

2.1. The Moroccan Atlantic coast

The Atlantic coast of Morocco is subject to intermittent upwellings of waters generally rich in nutrients and trace elements. This phenomenon is not constant along the coast, except for the Cape Spartel and the El Jadida–Safi sectors. The tide is semidiurnal, with variable amplitude (2.1 to 3.4 m). However, the tidal currents have been rarely studied. The Canaries Current, dominant at the surface, runs northeast to southwest at a rate of 0.5 to 9 $m.s^{-1}$, reaching its maximum force in the 20–50 m stratum. The direction of this current differs according to season (Fedoseev, 1970). The swell is always fairly heavy, with maxima of up to 9 m, and associated currents can disperse pollutant loads, ensuring their dissipation.

The Moroccan Atlantic coast is roughly straight. The dominant landscape along the coast consists of sand dunes, more or less transformed into sandstone, attaining 5 to 20 m in height and rarely covered by vegetation. Here and there, the coast is flat and rocky without cliffs, or lined with low, moderately high cliffs. These elements are suggestive of both an ‘African’ and a ‘Pacific’ coast (Bayed, 1982). The African type consists of large beaches, estuaries, and high or low cliffs that alternate along the coast. The Pacific type is characterised by old dunes, whose crests and sandstone ridges run parallel to the coast (Bayed, 1982).

2.2. Socio-economic activities and contamination sources

The Atlantic Larache–Safi area (*figure 1*) is the most important one in Morocco because of a high population and intensive industrial development. In fact, the Atlantic coastal belt contains 61% of the Moroccan population, with maximum urban density along the Kénitra–Safi area (Kaioua, 1991). The towns use the sea for final disposal of untreated wastewater, which contains a large and varied contaminant load. In 1990, the amount of wastewater evacuated by coastal urban areas along the area was estimated at 370×10^6 m^3, and this figure may be expected to reach around 10^9 m^3 within 30 years (Kaimoussi, 1996). According to Benchekroun and Bouchama (1991), urban discharges from Moroccan coastal towns constitute 55% of the total amount. The rapid demographic growth

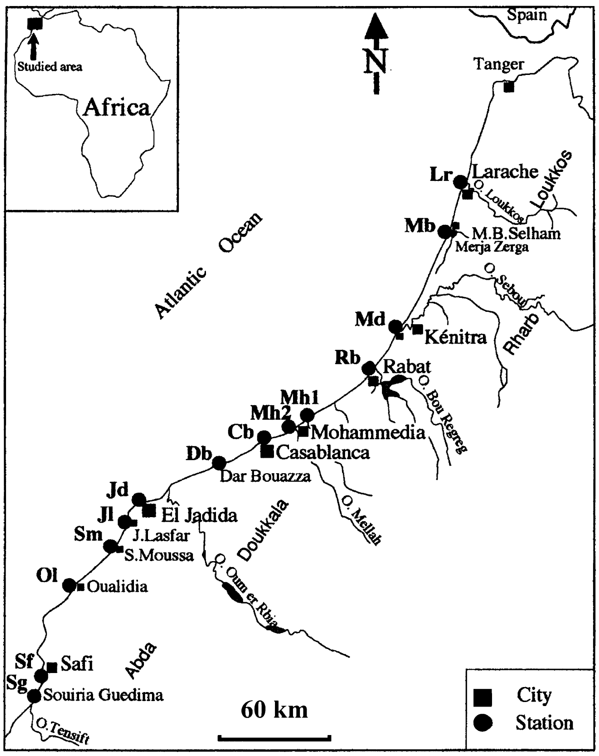

Figure 1. Location of coastal stations along the Moroccan Atlantic coast between Larache and Safi. Lr: Larache, Mb: Moulay Bou Selham, Md: Mehdiya (near Kénitra), Rb: Rabat, Mh1: Mohammedia Station 1, Mh2: Mohammedia station 2, Cb: Casablanca, Db: Dar Bouazza, Jd: El Jadida, Jl: Jorf Lasfar, Sm: Sidi Moussa, Ol: Oulidia, Sf: Safi, and Sg: Souiria Guedima.

along the coastal belt has generally been accompanied by a high concentration of economic and industrial activities. In fact, 80% of the industrial activities of Morocco are concentrated along the Atlantic coast. In 1990, this area accounted for more than 75% of firms and around 80% of jobs, mainly in the large cities. Seventy percent of the country's companies can be found between Kénitra and Safi, 85% of which exist in the administrative district of Casablanca. The dominant industries are of three main types: 1) metallurgical and electromechanical, 2) leather and textiles, and 3) chemical. Moreover, the Moroccan Atlantic seaboard concentrates 53% of tourism, and ensures 92% of maritime transport. In addition to purely commercial transport, fishing activities are concentrated on the Atlantic coast.

The Larache–Safi area includes four main plains with intensive farming activities, namely Loukkos, Rharb, Doukkala and Abda, which all have direct access to the Atlantic Ocean. This heavy agricultural development is associated with the use of increasing amounts of pesticides, fertilisers and other plant care products (biocides). According to several international organisations, such as the UNEP (United Nations Environmental Program) and WHO (World Health Organisation), the exaggerated and uncontrolled use of these substances can lead to chemical pollution, including cadmium, mercury and copper.

It is evident that each of these activities contributes directly or indirectly to the contamination of the marine environment with various chemical substances, including metals. The present study investigated the state of nutrient and metal contamination of the Moroccan Atlantic coast, using water analyses and the mussel (*Mytilus galloprovincialis*) as a quantitative bioindicator.

3. MATERIALS AND METHODS

3.1. Study stations

Fourteen coastal sites were chosen between Larache and Safi (*figure 1*) in order to take into account the influences of natural and anthropogenic characteristics, namely, hydrographical influences (upwelling, lagoons and estuaries) and urban, agricultural and industrial discharges. Details are given in section 2.2.

3.2. Samples

Water and mussel samples were obtained at low tide during the period 1993 to 1997. Water samples for physicochemical analyses were collected in polyethylene flasks previously rinsed with distilled water.

Mussels were collected by hand at the mean low water of neap tides on rocky substratum. Fifty adult individuals (around 5 cm in size) were sorted on the field. They were then cleaned and stored in polyethylene flasks containing water from the sampling site and kept in a cooler (+4°C).

3.3. Analyses

Seven physicochemical parameters were analysed in water. Temperature, total salinity (Model 33 YSI salinometer) and pH (ATC Piccolo HI 1280 pHmeter) were measured in situ. Dissolved oxygen was measured according to the protocol of Winkler adapted to seawater. Suspended particulate matter (SPM) was estimated by differential weighing before and after filtration (Whatman 0.7 µm) in a determined volume of water. Nitrate and phosphate analyses were performed according to the methodology of Aminot and Chaussepied (1983). Sampling bottles were rinsed in the laboratory with double distilled water and rinsed again with the sampled water just before collection.

After a period of purging in water from the sampling site, the mussels were transported to the laboratory in a cooler at +4°C. The soft parts of mussels were removed from their shells, thoroughly cleaned and drained for 2 h according to the protocol described by Claisse (1989). They were then homogenised using a grinder equipped with a stainless steel blade and freeze–dried for conservation until analyses. A subsample of 1 g was homogenised in a porcelain mortar before the mineralisation step. Mineralisation was performed with 4 mL Suprapur nitric acid (Merck) in polyethylene tubes at room temperature for 12 h and then at 90°C for 3 h in a water bath. After cooling, the digest was brought to 50 mL by addition of double distilled water.

Metals (Cu, Cd, Zn, and Mn) were measured by atomic absorption spectrophotometry (Perkin-Elmer, model 3100) equipped with a graphite furnace (HGA-600/700). The calculation of the coefficients of variation for each metal analysis, based on reproducibility of replicates analyses on one sample, gave the following relative coefficients of variation (i.e., standard deviation × 100 / average): Cu 11%; Cd 11%; Mn 13%; Zn 8%, i.e. a mean reproducibility of about 90%. The accuracy of the method was tested using a certified reference material obtained from the International Atomic Energy Agency (CRM No. MA-A-2/TM). The error brackets obtained for the various metals analysed ranged from 7 to 9%. The detection limits were 0.004, 0.13, 0.05, and 4 $\mu g \cdot g^{-1}$ (dry weight) for Cd, Cu, Mn, and Zn respectively.

3.4. Statistical analysis

Statistical analysis performed with the Bioméco programme (*Biométrie écologique*) concerned mainly the calculation of correlations between various parameters. Analysis of variance (ANOVA) with Microsoft Excel software (version 7.0) allowed comparison of metal concentrations in mussel tissues relative to site and season.

Table I. Interannual means and standard deviations for hydrological parameters at marine stations along the Larache–Safi area.

Site	Temperature (°C)	Salinity ($g \cdot kg^{-1}$)	pH	Oxygen ($mg \cdot L^{-1}$)	SPM ($mg \cdot L^{-1}$)	Phosphate ($\mu g \cdot L^{-1}$)	Nitrate ($\mu g \cdot L^{-1}$)
Lr	19.1 ± 3.9 (13.5–24.9)	28.9 ± 2.4 (23.9–32.2)	7.9 ± 0.3 (7.3–8.2)	7.3 ± 0.8 (5.7–8.2)	41 ± 10 (21–60)	76 ± 37 (23–160)	84 ± 123 (23–480)
Mb	18.9 ± 3.9 (14–25)	31.2 ± 2.3 (28–35.2)	7.9 ± 0.3 (7.4–8.3)	8.0 ± 1.1 (6.5–9.0)	43 ± 10 (33–65)	117 +115 (30–470)	77 ± 68 (24–240)
Md	19.7 ± 4.0 (14–27)	29.4 ± 1.9 (26–32.9)	7.7 ± 0.3 (7.2–8.1)	6.2 ± 1.5 (2.76–8.0)	69 ± 34 (44–183)	268 +127 (107–527)	171 ± 262 (40–950)
Rb	19.6 ± 4.1 (15–26)	32.2 ± 1.4 (30–34.2)	7.8 ± 0.2 (7.3–8.0)	6.7 ± 1.6 (3.3–8.4)	34 ± 10 (20–50)	334 ± 213 (116–941)	181 ± 269 (41–980)
Mh1	18.7 ± 3.7 (13–24)	32.4 +2.3 (25–34.5)	8.0 ± 0.2 (7.4–8.3)	6.9 ± 0.7 (5.3–8.0)	27 ± 5 (19–35)	144 ± 159 (22–468)	41 ± 27 (23–100)
Mh2	19.1 ± 3.5 (14–24.5)	27.9 ± 2.4 (24.1–32)	7.5 ± 0.4 (6.8–8.0)	4.4 ± 2.1 (0.0–7.6)	58 ± 13 (42–86)	675 ± 446 (230–1858)	187 ± 41 (49–198)
Cb	19.8 ± 3.9 (15–25.5)	30.0 ± 1.5 (27.9–33)	7.4 ± 0.4 (6.6–8.0)	4.8 ± 1.4 (1.05–6.2)	60 ± 8 (47–76)	273 ± 105 (116–520)	325 ± 350 (57–1042)
Db	18.1 ± 2.9 (13.5–22)	32.6 ± 2.3 (28.3–35.8)	8.1 ± 0.2 (7.6–8.2)	8.0 ± 0.2 (7.4–9.5)	30 ± 9 (19–52)	92 ± 15 (76–129)	69 ± 29 (29–120)
Jd	17.1 ± 3.6 (11–24)	30.5 ± 2.1 (26.4–34)	7.9 ± 0.1 (7.8–8.1)	5.5 ± 0.4 (4.2–6.8)	40 ± 4 (31–46)	600 ± 53 (300–1500)	263 ± 63 (199–342)
Jl	19.5 ± 3.6 (13.5–25.5)	32.7 ± 1.4 (28–35.2)	7.7 ± 0.1 (7.2–8.0)	6.0 ± 0.7 (5.0–7.8)	65 ± 7 (50–6)	8910 ± 11 (640–19 830)	209 ± 101 (34–470)
Sm	18 ± 3.5 (12.7–22.5)	32.9 ± 1.5 (30.5–36)	7.9 ± 0.2 (7.5–8.1)	6.6 ± 0.9 (4.4–7.6)	35 ± 13 (15–53)	409 ± 382 (185–1710)	130 ± 78 (31–310)
Ol	17.3 ± 3.4 (11.5–21)	31.9 ± 1.8 (27.5–35)	7.9 ± 0.1 (7.8–8.1)	6.9 ± 0.8 (4.5–8.0)	36 ± 9 (20.5–49)	250 ± 120 (130–590)	171 ± 139 (51–581)
Sf	17 ± 3.15 (12–21)	33.5 ± 1.1 (32–36)	7.9 ± 0.1 (7.7–8.1)	6.3 ± 0.8 (5.3–8.0)	57 ± 6 (49–67)	4460 ± 514 (850–19 220)	108 ± 100 (24–440)
Sg	17.1 ± 2.9 (12–20.5)	33.0 ± 1.7 (27.9–35)	7.9 ± 0.2 (7.3–8.1)	6.9 ± 0.7 (5.7–7.7)	47 ± 6 (36–61)	850 ± 0.5 (290–1680)	136 ± 121 (36–440)

Values were calculated from 16 determinations between 1993 and 1997. In parentheses: range throughout the study period. SPM: suspended particulate matter. Lr: Larache, Mb: Moulay Bou Selham, Md: Mehdiya (near Kénitra), Rb: Rabat, Mh1: Mohammedia Station 1, Mh2: Mohammedia station 2, Cb: Casablanca, Db: Dar Bouazza, Jd: El Jadida, Jl: Jorf Lasfar, Sm: Sidi Moussa, Ol: Oulidia, Sf: Safi and Sg: Souiria Guedima.

4. RESULTS

4.1. Hydrological parameters

The means for hydrological parameters, cumulated throughout the study period (1993–1997), are given in *table I*. Water temperature showed slight seasonal variations all along the Larache–Safi area. Variations of total salinity according to station were generally of low amplitude, although stations near river estuaries recorded a moderate decrease, notably at Larache, Mehdiya and Mohammedia (respectively near the mouths of the Loukkos, oued Sebou and oued Mellah). These results indicate the extent of inputs from continental waters into the ocean. A slight decrease in salinity was also observed at El Jadida and to a lesser extent at Casablanca, apparently due to the large quantity of effluents entering the sea near these stations. The mean pH values were close to 8, indicating the buffer effect of oceanic waters.

The interannual means for dissolved oxygen in water showed low levels in stations receiving untreated urban and industrial wastewater, particularly Mohammedia, Casablanca, El Jadida, Jorf Lasfar and Safi. Hypoxia and even anoxia were observed at Mohammedia and Casablanca (*table I*). The water at the other stations appeared to be well oxygenated. Parallel to a decrease in the oxygenation rate of waters from urban sites, suspended particulate matter (SPM) showed marked peaks, clearly indicating the input of particles at Mohammedia, Casablanca, Mehdiya, Jorf Lasfar and Safi. Likewise, stations receiving urban wastewater showed high nitrate concentrations, especially at the level of Mehdiya, Rabat, Mohammedia, Casablanca and El Jadida (*table I*). The average concentrations at Oualidia, Sidi Moussa and Souiria Guedima stations were fairly high, whereas those recorded at Larache and Moulay Bou Selham were in the low range. Spatial variations in mean phosphate concentrations showed two marked peaks for Jorf Lasfar (8.9 mg·L^{-1}) and Safi (4.5 mg·L^{-1}). The mean interannual concentrations at the other stations were high, due essentially to urban discharges at Rabat and Mehdiya, urban and industrial discharges at Mohammedia, Casablanca and El Jadida, and agricultural inputs at Moulay Bou Selham, Oualidia, Sidi Moussa and Souiria Guedima.

4.2. Metal analyses in *Mytilus galloprovincialis*

The mean metal concentrations cumulated throughout the study period (1993–1997) are reported in *table II*. The ANOVA on the basis of these concentrations (*table III*) showed significant variations according to station and sampling season.

Table II. Interannual means and standard deviations for metal concentrations in the mussel *Mytilus galloprovincialis* along the Larache-Safi area.

Site	Cu	Cd	Mn	Zn
Lr	10.6 ± 3.9 (5–18)	0.5 ± 1.0 (nd–0.9)	12.5 ± 5.2 (5.5–25.5)	201 ± 113 (175–533)
Mb	8.5 ± 3.5 (4–15.8)	0.4 ± 0.2 (nd–0.7)	13.3 ± 4.4 (5.4–18.5)	117 ± 34 (72–184)
Md	10.4 ± 3.4 (6.9–19.6)	0.7 ± 0.3 (nd–1.9)	19.1 ± 3.8 (14.4–27.9)	236 ± 72 (84–325)
Rb	13.4 ± 4.5 (8.1–23.1)	0.6 ± 0.3 (nd–1.5)	14.4 ± 4.3 (7.9–20.2)	256 ± 45 (186–317)
Mh1	6.9 ± 1.2 (5.2–9.5)	0.4 ± 0.3 (nd–0.7)	9.6 ± 2.7 (6.1–16.1)	241 ± 94 (96–401)
Mh2	14.4 ± 3.9 (10–24.2)	1.1 ± 0.9 (nd–2)	15.0 ± 2.8 (10.9–20)	338 ± 94 (265–645)
Cb	18.1 ± 9.5 (9.5–40.1)	1.9 ± 1.0 (0.5–3)	8.8 ± 2.2 (5.9–13.2)	379 ± 66 (250–481)
Db	4.9 ± 1.2 (2.9–7)	0.2 ± 0.3 (nd–0.5)	7.0 ± 1.6 (5.2–10.3)	152 ± 25 (116–195)
Jd	6.7 ± 3.9 (4–12.5)	1.0 ± 0.8 (0.1–2.9)	6.3 ± 0.7 (5.5–7)	186 ± 142 (110–398)
Jl	73.5 ± 42.0 (17–209)	8 ± 5.9 (3.5–15.5)	7.4 ± 2.4 (3.2–12.4)	267 ± 70 (189–456)
Sm	12.0 ± 3.8 (5.6–16.7)	1.4 ± 1.1 (nd–2.5)	13.0 ± 3.2 (8.8–18.5)	301 ± 151 (85–545)
Ol	7.5 ± 2.6 (4.3–12)	0.9 ± 0.7 (nd–1.8)	11.6 ± 4.5 (6.3–20)	202 ± 66 (96–296)
Sf	24.7 ± 5.5 (12.5–39.7)	7 ± 3.8 (3.3–12.4)	8.5 ± 4.4 (3.7–17.1)	290 ± 59 (177–370)
Sg	7.9 ± 3.6 (4.6–18)	1.5 ± 1.9 (0.5–3.8)	8.3 ± 2 7 (4–12.7)	277 ± 80 (67–371)

Values (in micrograms per gram, dry weight) were calculated from 16 determinations between 1993 and 1997. In brackets: minima and maxima throughout the study period. nd: not detected (< detection limit). Lr: Larache, Mb: Moulay Bousselham, Md: Mehdiya (near Kénitra), Rb: Rabat, Mh1: Mohammedia Station 1, Mh2: Mohammedia station 2, Cb: Casablanca, Db: Dar Bouazza, Jd: El Jadida, Jl: Jorf Lasfar, Sm: Sidi Moussa, Ol: Oulidia, Sf: Safi and Sg: Souiria Guedima.

Table III. *F* values for the analysis of variance (ANOVA) on metal concentrations in the soft tissue of *Mytilus galloprovincialis*.

Factor	Cu	Cd	Mn	Zn
Season	3.03***	1.94*	10.12***	6.35***
Site	27.91**	54.66***	19.41***	11.67**

Results show that space (site) and time (season) discriminate relatively to Cu, Cd, Mn and Zn concentrations. Asterisks refer to significance levels: * $P < 0.05$; ** $P < 0.01$; *** $P < 0.001$.

4.2.1. Spatial variations

Spatial variations in mean Cu concentrations were characterised by three clearly distinct peaks of variable intensity at Jorf Lasfar and Safi (which showed higher levels than other sites). Geographical variations in mean Cd concentrations were quite close to those for Cu. The peaks at the Jorf Lasfar and Safi sites were markedly above those of the other stations. The nature of activities nearby (processing of phosphate ores) suggests that inputs came from industrial and mining operations. The lowest Cd levels were recorded at Dar Bouazza, followed very closely by those at Moulay Bou Selham, Larache and north Mohammedia. Mussels from Mohammedia–Casablanca showed the highest Zn concentrations, followed by those collected at Jorf Lasfar, Sidi Moussa, Safi and Souiria Guedima. Similar levels were observed at Mehdiya and north Mohammedia, and lower levels at Moulay Bou Selham and Dar Bouazza. The highest mean Mn concentrations were observed at stations near estuaries and lagoons, especially Mehdiya, Mohammedia-town, Rabat, Moulay Bou Selham and Sidi Moussa. However, Mn concentrations were significant at Larache, the entry to oued Loukkos, and Oualidia. These results suggest that the sources of Mn are from riverine inputs.

4.2.2. Seasonal variations

The variations in seasonal metal concentrations in tissues of the mussel *M. galloprovincialis* along the Atlantic Larache–Safi area are indicated in *figure 2*, with distinct bars for the means obtained for each season. The seasonal effect is testified by the results of an analysis of variance (*table III*), which shows highly significant *F* coefficients for all metals studied. The amplitude of the seasonal variations is usually less than 20% but may reach a factor of 2. Mn and Zn concentrations for most stations were maximal in spring and minimal in winter (*figure 2*). Though the minima for Cu and Cd were also observed in winter.

5. DISCUSSION

5.1. Quality of marine waters

The spatiotemporal variations of various parameters, notably nutrient salts, dissolved oxygen in water

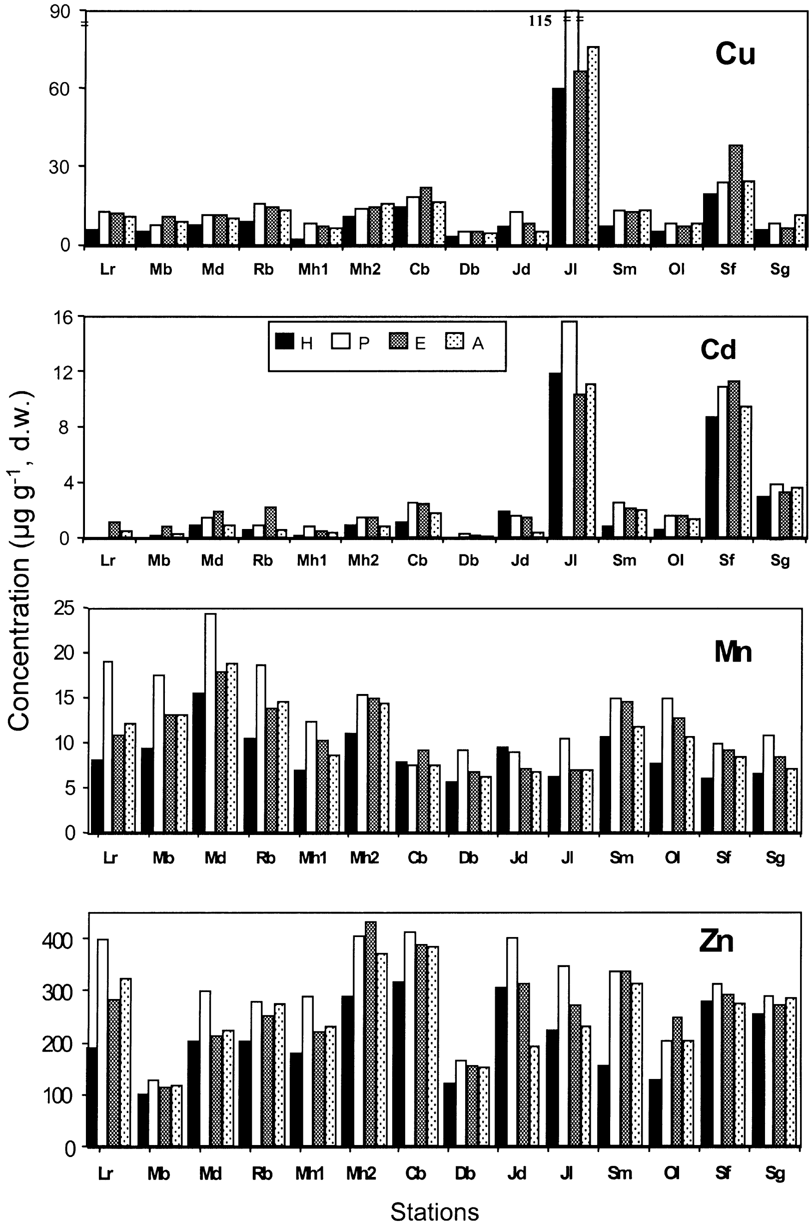

Figure 2. Seasonal variations in metal concentrations in the mussel *Mytilus galloprovincialis* on the Moroccan Atlantic coast between Larache and Safi. Lr: Larache, Mb: Moulay Bousselham, Md: Mehdiya (near Kénitra), Rb: Rabat, Mh1: Mohammedia Station 1, Mh2: Mohammedia station 2, Cb: Casablanca, Db: Dar Bouazza, Jd: El Jadida, Jl: Jorf Lasfar, Sm: Sidi Moussa, Ol: Oulidia, Sf: Safi and Sg: Souiria Guedima. H: winter, P: spring, E: summer, and A: autumn.

and SPM, are indicative of the quality of marine waters (Levitus et al., 1993; Fanning, 1992; Cheggour et al., 1999).

In our study, water temperature differed little from one site to another, whereas variations in total salinity showed a significant decrease near river estuaries, indicating the presence of continental inputs, especially in the rainy season. The relatively marked drop in water temperature between El Jadida and Safi showed that cold currents due to upwelling were predominant in these areas. The large temperature rise recorded at Jorf Lasfar (19.5°C) can be attributed to the discharge of hot industrial wastewater from phosphate processing complexes and a power plant.

The concentrations of nutrients were within the natural concentration range at Larache and Moulay Bou Selham in the north, and at Sidi Moussa and Oualidia in the centre. However, their seasonal cycles (with high values in winter and low values in summer) were disturbed in urban centres such as Mohammedia–Casablanca because of considerable human activity. Phosphate concentrations were particularly high at Jorf Lasfar and Safi, relative to phosphate-rich effluents from industrial complexes processing phosphates. These levels largely exceeded the means found for surface waters of the Atlantic Ocean (Fanning, 1992; Wiesenburg, 1988). High concentrations have also been reported for the Seine Bay (France), which drains a heavily industrialised area, notably with phosphate processing plants (Anon., 1990). The average concentrations calculated for Oualidia and Sidi Moussa reflect agricultural practices in these two regions. The nitrates used are recycled by tidal effects through lagoon systems before reaching the sea. The low values recorded at Larache and Moulay Bou Selham were due to low contaminant flux and high marine dilution. The dilution seems more effective at Moulay Bou Selham because of the large size of the lagoon (35 km^2), which drains highly developed agricultural land in the region. This would account for the low nitrate level found here compared to those of the Oualidia and Sidi Moussa sites where adjacent lagoons are of much smaller size (6 and 7 km^2). The nitrates detected near large cities suggest urban origin. However, an industrial source was likely, especially at Casablanca, relative to the activity of fertiliser plants at Aïn Sebaa and Jorf Lasfar. The results for El Jadida are consistent with the findings by Kaimoussi (1996), who studied nitrate variations at stations receiving urban sewages. These findings were confirmed by SPM values, whose high peaks at urban sites were clearly indicative of an input of particles due to urban effluents. The cases of Mohammedia, Casablanca, Mehdiya, Jorf Lasfar and Safi constitute quite significant examples (*table I*).

Dissolved oxygen also showed low levels at these same sites. The values observed (*table I*) are consistent with the quantitative importance of the pollutant loads in effluents at the sites considered (Benbrahim et al., 1997). These discharges were rich in organic detritus whose bacterial oxidation reduced dissolved oxygen in the environment. Nonetheless, in most of the sites the levels observed did not appear to be critical for marine life because of the continual hydrodynamic mixing of coastal waters, which favours the dilution of the organic matter load and allows rapid reoxygenation of the water. The level of dissolved oxygen recorded at Mehdiya reflects the effect of inputs from the oued Sebou, which conveys wastewater from the town of Kénitra and residues from port activities at both Kénitra and Mehdiya. The values recorded at the coastal stations of Oualidia and Sidi Moussa were related to local organic inputs originating mainly in the two lagoons and evacuated into the sea during their emptying at ebb tide.

Depending on the site, the results for pH indicated a greater or lesser enrichment of coastal waters with various forms of detritus emanating mainly from adjacent urban areas. This enrichment was significant at stations receiving discharges of urban and/or industrial wastewater either directly or via river estuaries. This accounts for the 'lower' pH (compared to marine pH) at Mehdiya, Rabat, Casablanca, Safi and Jorf Lasfar.

5.2. Metal contamination of the mussel *M. galloprovincialis*

5.2.1. Reasons for variations in metal concentrations

The variations in metal concentrations in mussels depended on several parameters. Coastal environmental factors such as temperature and salinity (abiotic factors) were taken into account as well as biological variables peculiar to the species, particularly those governing reproductive activity (biotic factors) (Mance, 1987; Cossa, 1989; Langston and Spence, 1995). In fact, it is difficult in a natural environment to sort the role played by each of these factors in the overall assessment of bioconcentrations because of their concomitant action.

5.2.1.1. Abiotic factors

The metal concentrations recorded along the Larache–Safi area clearly showed that mussel populations exposed to urban and industrial discharges were more contaminated with metal than those at some distance from these sources. High concentrations of Cu, Zn and Cd are common in urban effluents (Guillaud and Romana, 1991). In addition, Cd contamination cases have been observed particularly in industrial or agricultural discharges related to the exploitation, processing or land disposal of phosphate minerals (fertiliser and refuse) (Chiffoleau et al., 1994). The significantly negative correlations of these three metals with dissolved oxygen ($R_{Zn} = -0.51$, $P < 0.05$; $R_{Cu} = -0.53$, $P < 0.05$; and $R_{Cd} = -0.58$, $P < 0.01$) tend to confirm these hypotheses. Wastewater is a source of metals and organic matter, which cause an impoverishment of dissolved oxygen. This heavy load has been detected by analysis of effluents, notably at Casablanca (Benbrahim et al., 1997), El Jadida (Kaimoussi, 1996) and Rabat (Lemine, 1993).

Significant positive correlations were found between phosphate and nitrate on the one hand and Cd on the other (respectively, $R = 0.61$, $P < 0.01$; and $R = 0.57$, $P < 0.01$), and between Cu and phosphates ($R = 0.51$, $P < 0.05$). These relationships support the idea that Cd (and even Cu) inputs are of agricultural origin, resulting from the leaching of cultivated fields previously enriched with fertilisers and products for plant treatments that contain metals. However, an industrial origin is probably superimposed at Jorf Lasfar and Safi, owing to discharges from chemical complexes that process phosphates.

Manganese concentrations increased markedly after the winter, in conjunction with rainfall maxima and an associated drop in salinity and temperature. The significant negative correlations of Mn with salinity on the one hand ($R = -0.61$, $P < 0.01$) and temperature on the other ($R = -0.70$, $P < 0.01$) suggest a crustal origin for this metal. Alternatively, these correlations may also indicate the influence of hydrological parameters on the bioaccumulation of Mn within mussel tissues, as already reported by Mance (1987).

5.2.1.2. Biotic factors

Metal concentrations in mussel tissue showed seasonal cycles, with minima in winter. Depending on the station, these cycles were more or less developed and regular. This is in agreement with the results of Kaimoussi (1996) for the El Jadida region, where a marked spring peak was observed for several metals in soft tissues. In the Rabat–Mohammedia region, El Hraiki (1992) reported significant variations of Cu and Cd in the same mussel species and noted that concentrations were maximal in the spring. Both authors attributed these variations to the reproductive cycle of the species. Moreover, Asso (1984), in a study of another bibalve (*Perna perna*) in the Bay of Algiers, noted that several metals had an autumn maximum and a spring minimum, probably due to gametogenetic phenomena. Winter maxima and high seasonal amplitudes have been often observed in metal concentrations of *Mytilus* species from European coasts (e.g., Claisse, 1989). This dissimilarity may be due to differences in the biological cycle of the species (a divergence in time and individual phasing) related to latitudinal differences as a result of the various temperature cycles.

5.2.2. Origin and distribution of metals

Phosphogypsum discharges from local chemical complexes were responsible for much higher Cd and Cu concentrations in mussels at Jorf Lasfar and Safi than at other coastal stations (Cheggour et al., 1999). In fact, impurities rich in metals (notably Cu and Cd) have been frequently detected in phosphate concentrates produced in different regions of the world (Cossa and Lassus, 1989; Maxon and Vonkeman, 1992). Not surprisingly, the concentrations of these metals in mussels were markedly lower at other Moroccan sites where this type of industry is not present, i.e. the Rabat region (El Hraiki, 1992) and north of Larache (Idrissi et al., 1994). However, the relatively high Cu and Cd levels found at Mohammedia, Casablanca, Mehdiya and El Jadida are indicative of the role of urban and industrial activities in generating metal pollution. El Hraiki (1992) observed similar Cd concentrations in mussels from the Mohammedia region, in agreement with reports for North Atlantic mussels (ICES, 1980). Cadmium concentrations recorded at sites ‘not primarily industrial’ were generally lower, but still significant, as at Oualidia and Sidi Moussa on the central Atlantic coast and to a lesser extent at Larache and Moulay Bou Selham on the north coast. These concentrations could relate to the highly developed agricultural activities in these regions, which use large amounts of fertiliser and plant care products. Although Cd and secondarily Cu are minor constituents of these products, the large amounts used in intensive agriculture would contribute to their accumulation in the environment and their bioconcentration by organisms. It is also possible that the upwelling of waters rich in trace elements was a contributing factor (Bruland and Francks, 1983), notably at Oualidia and Sidi Moussa where this phenomenon is nearly constant. Measurements of the Cd flux in upwelling zones of the Atlantic and Pacific Oceans have indicated a marked increase in this metal, compared to areas some distance away (e.g., Cossa and Lassus, 1989).

Despite the magnitude of urban and industrial liquid–solid discharges and inputs from agricultural activities, which would seem to account for most of the Cd and Cu concentrations found in mussel tissues, atmospheric emissions cannot be entirely ruled out. Although this factor has not been thoroughly studied, Nriagu and Pacyna (1988) estimated that worldwide atmospheric emissions of Cd from anthropogenic activities represent several thousand tons (between 3 and 12) per year, mainly from industrial sources. For the Western Mediterranean, Migon et al. (1991) estimated annual atmospheric fluxes (in thousands of tons) at 0.14 to 0.17 for Cd and 1.5 for Cu.

The Zn concentrations recorded in mussels indicate the dominance of urban and industrial sources all along the Larache–Safi area. Levels were particularly high in the Mohammedia–Casablanca region where wastewater discharge outlets are large. Mussels also had relatively high Zn concentrations at the Rabat and Mehdiya stations, which receive discharges from large urban sewers as well as inputs from the Bou Regreg and Sebou estuaries respectively (*figure 1*).

A crustal origin was more apparent for Mn. The tissue concentrations of this metal in mussels were quite elevated in stations close to the mouths of estuaries and lagoons and remarkably low in typical urban areas distant from these points. These results are in close agreement with other Moroccan studies indicating that Mn is an excellent tracer of continental inputs into aquatic systems produced by the breaking up of rocks and soils of surrounding watersheds (Carruesco, 1978; Texier et al., 1994).

The present results for Morocco are concordant with those of other studies performed for areas along the Atlantic coast. For the El Jadida region, Kaimoussi (1996) found high metal concentrations in surface sediments, algae and mussels sampled near urban and industrial wastewater sewers. Echab et al. (1996) observed high metal concentrations around effluents from the town of Mohammedia, both in marine waters and mussels. The latter, collected on the Casablanca coast, were heavily contaminated with metals, especially at stations receiving wastewater (Cheggour et al., 1996). These results were confirmed by ecological studies along the Casablanca coast, which showed a significant decrease in the diversity of intertidal macrobenthos near effluents of anthropogenic origin (Chafik et al., 1999). Physicochemical analyses of these effluents, performed regularly by the National Institute of Halieutic Research (INRH) in Casablanca showed high organic and unusually high metal concentrations (Benbrahim et al., 1997). Similarly, our results at Safi are in complete agreement with those of Idrissi-Aatouf et al. (1996), who reported marked contamination of mussels (and certain gastropods) in close relation to urban and industrial effluents.

At an international level, the studies of Lauenstein and Dolvin (1992) conducted along American coasts reported significantly high metal concentrations in *M. edulis*, which were attributed mainly to human activities. For the same species in the Baltic Sea, Broman et al. (1991) observed relatively high Cd and Zn levels, and described a north–south gradient determined by salinity variations as well as by anthropogenic inputs into the marine environment. The monitoring of chemical contamination along French coasts over a period of several years, using *Mytilus* sp. as a quantitative biological indicator, has allowed the identification of certain 'hot spots' for anthropogenic activity, characterised by particularly elevated levels of some metals such as Cd (Claisse, 1989).

5.2.3. Comparison of the stations and the pollution gradient

To compare metal pollution levels in mussels along the Moroccan Atlantic coast, a metal contamination index Ic_{Sx} was calculated according to the equation:

$$Ic_{Sx} = Sx/M$$

where Sx is the mean concentration (in micrograms per gram, dry weight) for metal x at station S, and M the cumulative mean concentration (in micrograms per gram, dry weight) for the same metal at all stations. In the absence of any environmental standard, this index allows the degree of contamination in different localities to be compared with a regional average (in this case, that of Larache–Safi area). According to the results obtained (*table IV*), the following increasing contamination gradient for all the metals combined can be proposed: Dar Bouazza < (Oolidia, Sidi Moussa) < (Moulay Bou Selham, Larache) < Souiria < Mohammedia (station 1) < El Jadida < (Mehdiya, Rabat) < (Jorf Lasfar, Safi, Mohammedia (station 2, Casablanca).

The comparison of the present metal data in mussels with those of other localities in the world (*table V*) indicates that the level of contamination on Moroccan

Table IV. Metal contamination indices (Ic_{Sx}) for the mussel *Mytilus galloprovincialis* along the Moroccan Atlantic coast between Larache and Safi.

Site	Cu	Cd	Mn	Zn
Larache	0.68	0.24	1.12	0.81
Moulay Bou Selham	0.54	0.11	1.20	0.48
Mehdiya	0.67	0.34	1.73	0.96
Rabat	0.85	0.21	1.30	1.04
Mohammedia 1	0.44	0.20	0.87	0.98
Mohammedia 2	0.91	0.55	1.36	1.38
Casablanca	1.15	0.98	0.80	1.54
Dar Bouazza	0.31	0.10	0.63	0.62
El Jadida	0.43	0.51	0.57	0.76
Jorf Lasfar	4.68	4.12	0.64	1.08
Sidi Moussa	0.76	0.71	1.18	1.22
Oualidia	0.48	0.50	1.05	0.82
Safi	1.60	3.60	0.77	1.18
Souiria Guedima	0.50	0.80	0.75	1.13

See text for definition of Ic_{Sx} (section 5.2.3).

Table V. Range of metal concentrations in soft tissue of mussels from different localities in the world.

Locality	Cu	Cd	Zn	Reference
Larache–Casablanca, Morocco	6.6–14.4	0.4–1.9	117–379	Present study[a]
Jorf Lasfar–Safi, Morocco	24.7–73.5	7.0–8.0	267–290	Present study[a]
Rabat, Morocco	2.4–12.2	0.13–1.5	50–344	Chafai-El Alaoui (1994)[a]
Mohammedia, Morocco	13.6–16.8	0.8–0.93	–	Echab et al. (1996)[a]
El Jadida, Morocco	4.4–14.7	nd–2.5	148–535	Kaimoussi (1996)[a]
Bay of Algiers, Algeria	10.8–15.7	0.54–1.8	133–243	Abada (1996)[b]
English Channel, France	3.8–16.5	0.4–8.8	53–318	Boutier (1982)[c]
Mediterranean, France	4.1–20.9	0.2–3.4	61–1336	Boutier (1982)[a]
Atlantic, France	–	0.1–36.2	–	Claisse (1989)[c]
Mediterranean, Italy	2.4–154	0.4–5.9	97–644	Abada (1996)[a]
Coast of Portugal	6.2–13.4	0.5–1.3	140–542	Coimbra et al. (1991)[c]
Atlantic, Spain	2–4	7–14	190–370	Manga (1980)[c]
South Devon, UK	0.2–17.3	0.8–36.2	16–634	Boalch et al. (1981)[c]
Baltic Sea	–	5–11.5	121–160	Broman (1991)[c]
Coast of Sweden	–	3.8	117	Broman (1991)[c]
Long Island, NY, USA	5.5–70	0.7–10	–	Turgeon et al. (1989)[c]
USA coasts	5.7–530	0.9–9.1	67–6 000	Lauenstein et al. (1990)[c]
Pacific, USA	3.5–19.1	0.8–10.5	51–260	Goldberg et al. (1983)[c]
Pacific, Hong Kong	0.1–1.4	8.5–278	77–164	Phillips (1985)[d]
Victoria, Australia	3–11	2–140	126–747	Manga (1980)[c]
Derwent, Australia	24	42	352	Manga (1980)[c]

Values are in micrograms per gram, dry weight. (a) *Mytilus galloprovincialis*; (b) *Perna perna*; (c) *Mytilus edulis;* (d) *Perna viridis*; nd: not detected.

coasts is still moderate, except for a few sites. This justifies that current monitoring systems and pollution control are necessary and should be reinforced. When the results are considered in terms of the maximum concentrations proposed by the Oslo and Paris Conventions and British guidelines for molluscs, the quality of *M. galloprovincialis* relative to metal contamination can be regarded as good at most stations on the Moroccan Atlantic coast. However, Cd concentrations at Jorf Lasfar and Safi are close to the current standards established by the European Community regulation (No. 194/97).

6. CONCLUSION

The water analyses along the Moroccan Atlantic coast between Larache and Safi showed some deterioration in quality (especially dissolved oxygen and nutrients), at stations affected by heavy urban and industrial activities. This is the case for Mohammedia–Casablanca, Jorf Lasfar and Safi areas. Coastal waters were of higher quality at sites with little or no human activity.

The Mussel Watch programme suggests a dominant source of metal contamination from urban and industrial activities, and less important inputs from continental weathering and agricultural origins. Metal concentrations distribution in mussels indicates hot spots near large urban and industrial centres, notably Mohammedia–Casablanca. In addition, the mussels of Jorf Lasfar and Safi exhibit high Cd concentrations as a result of their exposition to industrial effluents. Consequently, these sites need to be monitored quite closely. Localities relatively far from anthropogenic sources (e.g., Moulay Bou Selham, Sidi Moussa and Oualidia lagoons on the Atlantic coast), which have considerable potential for aquaculture, were affected very little by chemical contamination.

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