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# The impact of mining activities on the hypersaline Mar Menor lagoon

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ABSTRACT

Mar Menor lagoon is one of the largest hypersaline coastal lagoons in the Mediterranean Sea. The lagoon is threatened by environmental degradation due to the urban growth around it, the intensive agriculture in its watershed and the remains of mining activities in the nearby Cartagena-La Unión area. The present study aims at estimating the levels of zinc and lead in the water column, sediments and marine flora and fauna of Mar Menor lagoon. The ranges of the total concentrations in the water column were 9.36–28.7 μg/L for Zn, 1.61–12.6 μg/L for Pb and 3.01–29.70 μg/L for Mn. Zinc was mostly found in the dissolved phase (70%) whereas lead and manganese in the suspended phase (63–76%). Highest values were measured at stations closer to streams flowing trough drainage basins including agricultural zone and the mining remains. Concerning sediments, the values were very high especially at the southern part, indicating transport of suspended matter from currents. The metal content in the sediments is higher than the content in particulate matter and there is an increased bioavailability of Zn and Pb. Zinc and lead were measured in the thallus of four marine flora species. Maximum zinc concentration was measured in *Cymodosea nodosa* (132 μg/g) and maximum lead in *Acetabularia acetabulum* (93.5 μg/g) in samples from the southern lagoon. From the two benthic fauna species studied, it was observed that heavy metals accumulate mostly in the animal flesh. The highest concentration of Zn and Pd was measured in the flesh of the mollusk *Ostrea edullis* with values of 1,856 and 51.0 μg/g respectively. These values reveal the effect of heavy metal pollution in the lagoon ecosystem.

***Keywords:*** Zinc; Lead; Manganese; Ferrum; Mar Menor; Lagoon; Marine flora; Benthic fauna

## 1. Introduction

Lagoons are stretches of salt or brackish water in coastal areas that are separated from the sea usually by narrow beach barriers. They can be connected to the sea either constantly or periodically [1]. Lagoons occupy 13% of the world's coastline [2] and are autonomous dynamic systems of high productivity potential sharing common characteristics, such as frequent fluctuations in abiotic parameters. Temperature, salinity, pH, nutrients and heavy metals may alter either naturally or artificially. Human induced changes in nutrient and heavy metal concentrations have been observed in many lagoons worldwide, causing serious changes in the abundance and distribution of organisms [3].

The fate of heavy metals in the aquatic environment depends on biological activity and geochemical

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processes. Their intake by organisms via the food chain possibly results in bioaccumulation and biomagnification. On the other hand, metals are released during the degradation of organic matter [4] and may be transferred to the sediments through adsorption onto settling suspended matter. Moreover, metals can be desorbed from the particulate material or the sediments due to the variations of physicochemical parameters such as salinity, temperature, dissolved oxygen, pH [4,5].

Therefore, as also suggested by the message of the Global Day of Wetlands in 2008, which was 'Healthy Wetlands-Healthy People', heavy metal levels monitoring in lagoon environments is essential in order to assess and apply any necessary measures so as to avoid ecological degradation due to increased concentrations of heavy metals in the water and sediment. Hence, aquatic flora and fauna as well as humans may be protected against serious health implications of heavy metal pollution [1,6].

The present study aims at estimating the levels and distribution of zinc and lead in the water column, sediments and marine flora and fauna of Mar Menor lagoon which is affected by various anthropogenic activities.

## 2. Study area

Mar Menor lagoon is one of the largest hypersaline lagoons of the Mediterranean Sea, measuring about 13 × 15 × 21 km and having a mean depth of 2.5 m. It is constantly hypersaline, as the freshwater influx from the streams (ramblas) of Sierra de Cartagena, La Unión and the Campo de Cartagena areas is not enough to compensate for the high evaporation, especially during summer.

Mar Menor is separated from the Mediterranean by a narrow sandy coastal barrier called La Manga and is connected to it via one natural and two artificial channels. It is surrounded by urban areas, agricultural sites and salt marshes, with flora tolerant to salinity, drought and high temperatures. Moreover, Mar Menor is a shelter for many species of endemic and migratory birds and it is a Ramsar International Site since 1994. Five regions around it have been characterized as Sites of Community Importance, while it is protected by the Birds Directive 79/409/EEC and included in the NATURA 2000 Network.

The mountains of Cartagena and La Unión (Sierra Minera) areas are rich in iron, lead and zinc ores. Lead and zinc mining activities had started here from Roman times and continued until the 20th century, finally ceasing in 1991. There are yet a lot of mine tailings at these mountains, most of them exposed to rain and wind, that cause erosion and pollution problems. High zinc and lead concentrations have been determined in the water of the streams, in the soil of their mouths and in the salt marshes.

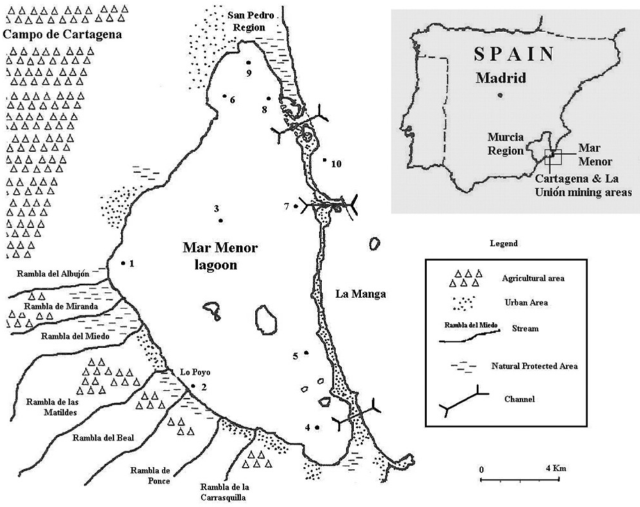


Fig. 1. The Mar Menor lagoon and its surrounding area, including sampling stations (originally from Conesa et al. [1].

The Campo de Cartagena is an agricultural region. Cultivation practices changed during the 1980s from extensive dry crops to irrigated ones, through the Tajo-Segura river diversion. A lot of greenhouses have also been set up in the area.

Nowadays, tourism is one of the main activities in the area and has led to the urbanization of La Manga. The population around the lagoon increases dramatically during summer.

It is due to all these reasons why the lagoon environment is threatened by eutrophication phenomena and by heavy metal pollution [1,3,7] (Fig. 1).

## 3. Materials and methods

Fieldwork took place during May 2007. The sampling stations were selected in such a way to cover all parts of the lagoon: the north (stations 6, 8, 9), the central (stations 1, 3, 7) and the south (stations 2, 4, 5). Station 10 was located outside the lagoon, in the Mediterranean Sea, in order to be used as a reference station. In particular, stations 1 and 2 are close to the mouths of the streams that flow through the mining and agricultural areas.

Water temperature, salinity, dissolved oxygen concentration and water depth were measured in-situ by portable equipment. Hydro-Bios polypropylene bottles were used for water sampling. Water samples were filtered sequentially through 8 μm and 0.45 μm Millipore nitrocellulose filters and stored in a refrigerator at 4 °C. All samples were handled in a laminar flow bench in order to avoid any contamination [8]. Filtered samples were pre-concentrated by passing through Chelex 100 resin columns. The dissolved metals were eluted from the resin by $HNO_3$ 2N. The filters carrying the suspended material were digested at 120 °C with concentrated nitric acid [9,10].

Divers collected surface sediment samples as well as samples of four species of marine flora (*Caulerpa prolifera*, *Cymodocea nodosa*, *Cystoseira mediterranea*, *Acetabularia acetabulum*) and of two benthic fauna species (*Ostrea edulis*, *Hexaplex trunculus*).

The sediment samples were stored at −20 °C, then freeze-dried and dry-sieved. The total metal content in sediments was determined after digestion of samples in concentrated $HNO_3$ - HF in PTFE beakers. Non-lattice held metals were extracted by 0.5N HCl [11–13].

The lagoon organism samples were transferred to the laboratory inside a portable fridge filled with ice. The samples were freeze-dried, homogenized, weighed and digested with concentrated nitric acid (65%) in PTFE beakers for 2 h at 140 °C [14].

Zn, Fe and Mn were determined by the use of a VARIAN SpectrAA 200 instrument (Flame AAS) and Pb was determined by the use of a VARIAN SpectrAA 640Z instrument (Graphite Furnace AAS) with Zeeman background correction [9]. Fe was measured only in the particulate matter, as it is not removed quantitatively from the samples by Chelex 100 resin. The methods have been tested in the laboratory for their accuracy and reproducibility using reference materials and replicates [15,16]. The relative standard deviations were: Dissolved Zn 4.6%, Particulate Zn 5.5%, Dissolved Pb 6.7% and Particulate Pb 5.9%.

## 4. Results and discussion

### 4.1. Physical parameters

The temperature of the lagoon water ranged between 26.4 and 28.1 °C and it was higher than that of the Mediterranean Sea (25.1 °C) at sampling station 10. The highest value was measured at station 9 whereas lower values were determined at stations 3, 4, 5 and 7. Due to the small water depths of the lagoon no thermocline was observed.

The salinity in the lagoon ranged between 42.6 and 44.0 psu with an average of 43.3 psu, 5.7 units higher than the sea value (37.6 psu.). The highest values were measured at stations 2, 4 and 5.

Table 1
Range of suspended particulate matter concentrations (mg/L)

| Sampling station | Fine fraction $0.45 < d < 8$ μm | Coarse fraction $d > 8$ μm |
|---|---|---|
| Mar Menor | 10.7–20.4 | 10.0–25.0 |
| Reference station | 7.6 | 10.0 |

The dissolved oxygen in the lagoon was 4.5–6.6 mg/L with an average value of 5.2 mg/L. The calculated oxygen saturation of the lagoon water was more than 80%, indicating its good oxygenation level. The, dissolved oxygen concentration in the Mediterranean coast was 8.2 mg/L, due to the lower temperature and salinity there.

As expected, the concentration of suspended matter in Mar Menor was higher than the concentration at the reference station (Table 1). Furthermore, the coarse fraction ($d > 8$ μm) was more than the fine fraction ($0.45 < d < 8$ μm), with values 55% and 45% respectively. The highest concentrations were measured at stations 1, 3 and 7 in the central part of the lagoon probably due to currents that transfer Mediterranean water into the lagoon, causing re-suspension of sediments [3].

### 4.2. Heavy metals

#### 4.2.1. Water column

In Table 2, the ranges of the concentrations of the studied metals, both in the dissolved and in the suspended phase, are presented together with those values derived from the reference sampling station 10 (located outside the lagoon).

As can be seen in Fig. 2, the total Zn, Pb and Mn concentrations in the lagoon waters were higher than those at the reference station. In contrast, the value of particulate Fe in the reference station was higher than those measured at stations 1, 6 and 8. The highest concentrations of all metals were measured at stations 2 and 5. The values at station 2 are attributed to its proximity to the mouth of the Rambla del Beal, winch is enriched in Zn (410 μg/L) and Pb (70 μg/L). In addition, this sampling station is in front of Lo Poyo salt marsh, where the soil concentration of Zn has been measured at 6,940 mg/kg and of Pb at 8,000 mg/kg [1]. The maximum concentrations of Mn and Fe were observed at station 5, as well as increased values of Zn and Pb, while at station 4 the highest concentration of Pb was measured.

Table 2
Ranges of concentration of Zn and Pb in the water column; average values are given in parenthesis

| | Zn μg/L | | Pb μg/L | |
|---|---|---|---|---|
| | Suspended | Dissolved | Suspended | Dissolved |
| Mar Menor | 1.72–15.6 (5.91) | 5.92–16.4 (12.0) | 0.61–8.73 (5.01) | 0.99–5.11 (2.38) |
| Reference | 4.24 | 7.24 | 0.94 | 0.58 |
| | Mn μg/L | | Fe μg/L | |
| | Suspended | Dissolved | Suspended | |
| Mar Menor | 2.06–25.7 (11.8) | 0.95–5.96 (3.68) | 11.3–167 (71.5) | |
| Reference | 0.70 | 0.46 | 30.4 | |

Taking into account the direction of currents in the lagoon, as described in Pérez-Ruzafa et al. [3], we consider that the heavy metals entering from the ramblas debouching at the southwest coast of the lagoon are transferred to the south part, i.e. at stations 4 and 5. In addition, these sampling points may be also affected by wastewater discharges from the urbanized area of La Manga.

At the northern part of the lagoon (stations 6, 8, 9) the local maximum values of all studied metals were observed at station 9. This area is shallow and hosts many wild animals and birds, which may induce re-suspension of the lagoon bottom sediment. Moreover, the industrial activity in the region of San Pedro may be considered as a potential source of heavy metals.

The similarity in the bahavior and concentrations distributions between Zn-Fe and Pb-Mn (Fig. 2) is confirmed also by the correlation coefficients ($R^2$), which for the total metals concentrations in the water column were 0.87 and 0.82, respectively.

Zinc was found mostly in the dissolved form (70%) and below the detection limit of the analytical method in the fine fraction of the suspended matter. On the other hand, only 37% of Pb was in the dissolved form, while its concentration in the coarse particulate matter reached 58% of the total. Likewise, the dissolved Mn was 29% of the total, while the coarse suspended fraction was 68%. Regarding the particulate Fe, its percentage in the coarse fraction was 93% of the total.

A comparison of pollutant concentrations in Mar Menor with relative values in other lagoons (worldwide)

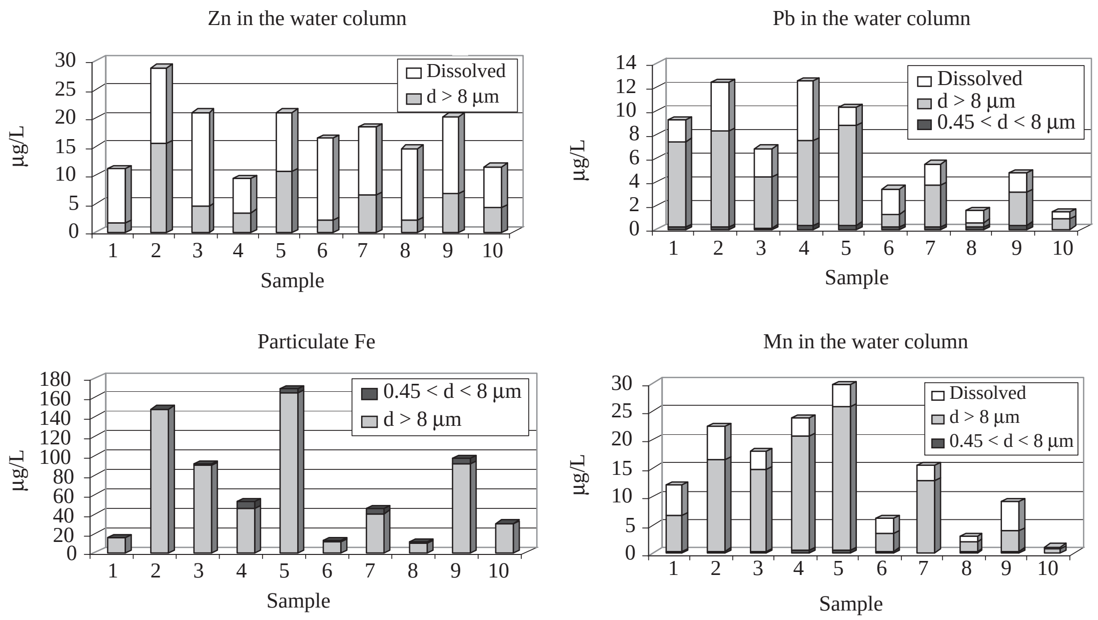


Fig. 2. Metal concentrations in sampling stations.

Table 3
Total heavy metal values (dissolved + total suspended) of the Mar Menor lagoon compared to other lagoons in the Mediterranean and the Gulf of Mexico

| Lagoon | Zn μg/L | Pb μg/L | Cited reference |
|---|---|---|---|
| Mar Menor (Spain) | 9.36–28.7 (17.9) | 1.61–12.6 (7.40) | Present study |
| Korissia (Greece) | 0.08–4.77 | 0.08–0.59 | [17] |
| Antinioti (Greece) | 2.63–7.72 | 0.09–0.25 | [18] |
| Moustos (Greece) | 0.07–3.44 | 0.04–0.55 | [19] |
| Alvarado (Mexico) | n.a. | 9.39–62.5 | [4] |
| Pom-Atasta (Mexico) | 9.29 | 69.1 | [5] |

*The values of Pb for Alvarado and Pom-Atasta lagoons refer to dissolved metals.

is presented in Table 3. Zinc and lead concentrations in Mar Menor were many times higher than those observed in the three Greek lagoons (Korissia, Antinioti and Moustos). Korissia, an intermittently hypersaline lagoon, and Antinioti, are located on Corfu Island and communicate with the open Ionian Sea; they are both affected by intensive agricultural activities taking place in their drainage basins and by wastewater discharges, especially during summer, due to tourism. Moustos lagoon is in southeastern Greece. On the other hand, heavy metal concentrations in Mar Menor are lower than those measured in the two Mexican lagoons (Alvarado and Pom-Atasta), which are located at the south part of the Gulf of Mexico. These lagoons are affected by human activities such as food processing, petroleum refining, sugar refining and agriculture.

#### 4.2.2. Sediments

The sediments in Mar Menor lagoon were mainly coarse-grained as the 63 μm < $d$ < 1 mm (silt and clay) fraction was on average 87% of the total. The carbonate content was also high (57%), being consistent with the increased levels of $CaCO_3$ found in the soils of the nearby Campo de Cartagena plain [1]. The carbonate content of the fine ($d$ < 63 μm) grain-size fraction, that comprised 13% of the total, was almost the same, i.e. 56%.

In Table 4 the ranges and median values of the concentrations of heavy metals in the two sediment fractions are presented, compared to the reference values of station 10. The w/w concentrations of the metals in the particulate matter are also included for comparison. The median values are used instead of the

Table 4
Ranges of heavy metal content of sediments and suspended matter (median values given in parenthesis)

| | | Zn mg/kg | | Pb mg/kg | |
|---|---|---|---|---|---|
| | | $d$ < 63 μm | 63 μm < $d$ < 1 mm | $d$ < 63 μm | 63 μm < $d$ < 1 mm |
| Mar Menor | Total | 90.8–3,372 (425) | 20.0–3,740 (818) | 57.5–2,977 (565) | 23.9–2,823 (605) |
| | Extractable | 57.2–3,317 (324) | 2.00–3,700 (744) | 36–2,975 (544) | 17.5–2,800 (672) |
| Ref. station | Total | 27 | 17 | 13 | 14 |
| | Extractable | 12.0 | 5.82 | 9.00 | 9.00 |
| | | Total suspended matter | | | |
| | Mar Menor | 24.2 – 593 (128) | | 14.0 – 315 (182) | |
| | Reference | 241 | | 53.7 | |
| | | Mn mg/kg | | Fe mg/kg | |
| | | $d$ < 63 μm | 63 μm < $d$ < 1 mm | $d$ < 63 μm | 63 μm < $d$ < 1 mm |
| Mar Menor | Total | 124–2,114 (390) | 97.6–1,638 (398) | 1,383 – 66,272 (18,714) | 1,195 – 50,785 (12,933) |
| | Extractable | 109–1,472 (268) | 88.4–1,467 (312) | 1,194–34,202 (2,748) | 698–39,754 (3,602) |
| Ref. station | Total | 193 | 100 | 4,286 | 1,660 |
| | Extractable | 128 | 87.6 | 1,032 | 542 |
| | | Total suspended matter | | | |
| | Mar Menor | 47.8–767 (166) | | 170 – 5,670 (1,959) | |
| | Reference | 40 | | 1,733 | |

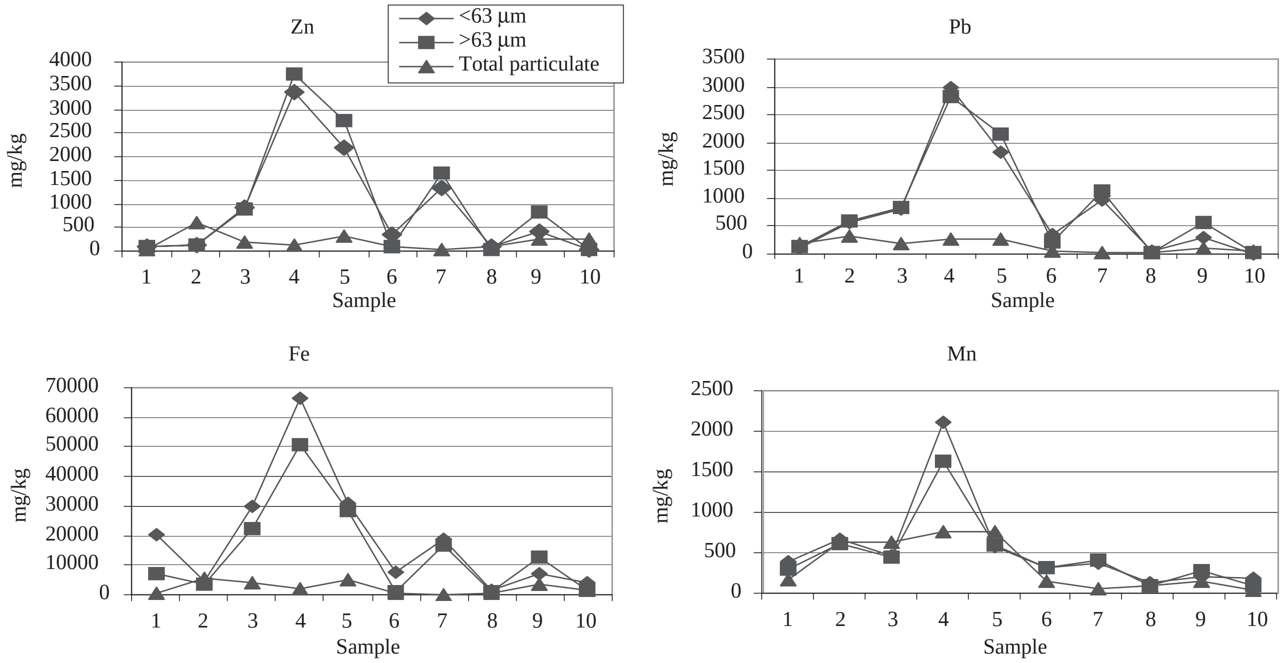


Fig. 3. Distribution of heavy metals in the lagoon bottom sediments.

average values, as the number of samples is low and the range of values is high.

The median heavy metal values in the lagoon were much higher than those in the Mediterranean Sea, reflecting the heavy metal load transferred from the Sierra Minera mining tailings. For Zn, the median value in the fine fraction was 48 times higher than that of the sea, and for Pb, it was 43 times higher for both grain-size sediment fractions. Concerning the non-pollutants Mn and Fe, the differences were lower and detected mainly in the coarse fraction.

The 0.5N HCl extractable concentrations of the studied metals are also presented in Table 4 and indicate increased mobility of zinc, lead and manganese. The median percentages of the extractable fraction for Zn and Pb were 83% and 94%, respectively, indicating the high degree of bioavailability of these pollutants.

The distribution of the heavy metals in the sediments samples of Mar Menor is shown in Fig. 3. The concentrations in the two grain-size fractions follow the same distribution pattern. The sampling stations 4 and 5 exhibit the highest values of heavy metal concentrations, whereas stations 7 and 9 comprise local maxima. Moreover, the values in stations 1 and 2, which are close to the streams that are the main pollution point sources, are lower than that of stations 4 and 5. It is, therefore, concluded that the south-eastern part of the lagoon is the most polluted, probably due to the current direction that transfers the pollutants there and to the high-level of urbanization of the surrounding area.

Furthermore, as shown in Fig. 3 and in Table 4, the heavy metal content of the suspended mater was much lower than that of the bottom sediments of the lagoon, while in most cases the values were close to those of the Mediterranean sediment (station 10). Taking into account that the mining activities have ceased for almost three decades and that the opening of artificial channels had improved the connection of Mar Menor with the Mediterranean it can be assumed that an increased part of the suspended material in the lagoon is derived from the open sea; this eventually will cover the heavily polluted bottom sediment with less polluted sediment.

Table 5 presents zinc and lead sediment concentrations in other lagoons worldwide. Zinc values in Mar Menor are comparable only to those of Szczecin & Gdansk lagoons (in Poland) and to Stettin lagoon (in Germany), as they are also affected by mining activities. The other lagoons are influenced primarily by agricultural and tourist activities in their drainage basin. It is also worth mentioning, that the bottom sediments of Mar Menor are, at least locally, heavily polluted by lead, as they have the largest concentration range.

### 4.3. Marine organisms

In Table 6 are presented the average concentrations of zinc and lead, measured in the thallae of marine plants. Zinc was the most abundant metal in *Cymodosea nodosa*, while lead in *Acetabularia acetabulum*.

Table 5
Zn and Pb sediment concentrations in other lagoons

| Lagoon | Zn μg/g | Pb μg/g | Reference |
|---|---|---|---|
| Mar Menor* | 20.0–3,740 (818) | 23.9–2,823 (605) | Present study |
| Korissia (Greece)* | 28.1–74.3 (59.3) | 20.9–56.7 (42.4) | [17] |
| Antinioti (Greece) | 5.4–122 | 4.0–53.0 | [18] |
| Moustos (Greece) | 106–142 | 20.6–22.5 | [19] |
| Nador (Morocco) | 40–212 | <73 | [20] |
| Sidi Moussa (Morocco) | 19–73 | | [21] |
| Edku (Egypt) | 0.4–11.3 | 4.2–7.3 | [22] |
| Fusaro (Italy) | 6.6–57.7 | 17–50 | [23] |
| Vistula (Poland) | 76–137 | 24–80.5 | [24] |
| Szczecin&Gdansk (Poland) | 166–1470 | 40–180 | [25] |
| Stettin (Germany) | 534–981 | 103–161 | [26] |
| Pom-Atasta (Mexico) | 19.0–74.4 | 1.45–23.8 | [5] |
| Great Astrolabe (Fiji Islands) | 10–164 | 4.0–17 | [27] |

* 63 μm < d < 1mm, grain-size fraction

It is obvious (Fig. 4) that the highest concentrations of zinc were found in the marine flora, located at sampling stations 2, 4 and 5, as well as at station 9. For lead, the highest values were measured at stations 2, 3, 4 and 5. The samples from the reference station 10 exhibited the lowest values.

These findings are in accordance with the heavy metal distribution in the lagoon sediments, as far as the sampling stations at the south part are concerned. The results at stations 2 and 4 are possibly related to the high Zn and Pb concentrations in the water column, as it is well known that submerged macrophytes are capable of feeding not only from their rhizomes but also from their thallae.

From the two benthic fauna species studied, the higher values of heavy metal concentrations were measured in the flesh of the mostly edible *Ostrea edulis*. Zinc reached 1,856 μg/g and lead 51.0 μg/g; these values, compared to others measured in the same species [28], indicate possible biomagnification of the two heavy metals.

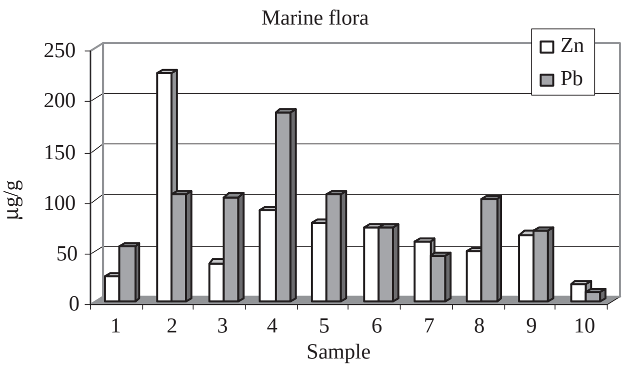


Fig. 4. Lead and zinc concentrations in marine flora species at the sampling stations.

Table 6
Average heavy metal concentrations in the plant thallus

| | Cp | Cn | Cm | Aa |
|---|---|---|---|---|
| Zn μg/g | 51.6 | 132.0 | 83.7 | 108.0 |
| Pb μg/g | 90.6 | 60.7 | 61.6 | 93.5 |

Cp: *Caulerpa prolifera*
Cn: *Cymodocea nodosa*
Cm: *Cystoseira mediterranea*
Aa: *Acetabularia acetabulum*

## 5. Conclusions

Mar Menor lagoon appeared to be affected significantly by heavy metal pollution. It is mainly due to the transfer of polluted water from the mining remains of the Cartagena-La Union area. The water circulation seems to play an important role in the distribution of the pollution and, consequently, in the influence on aquatic organisms.

The most affected part of the lagoon is its southeastern area, where some principal ramblas debouch and currents transfer run-off inputs from the southwestern coast. The highest concentrations of zinc and lead were measured there both in the water column and in the sediments. Moreover, it was found that the most of the Zn and Pb in the sediment can be bioavailable. Yet, the heavy metal content of the particulate matter was much lower than that of the sediments, with concentrations close to those of the Mediterranean Sea bottom sediment. This is probably the result of the artificial channels that connect the lagoon to the open sea together with the termination of mining activities. Hence, the deposition of these marine particles will eventually lead to the decrease of pollution levels in lagoon sediments.

The studied marine flora and fauna species exhibited higher Zn and Pb concentrations in the central part of the lagoon, as well as close to the mouths of the streams.

The findings of this study indicate that there is a need for prompt measures to be taken, in order to decrease the heavy metal inflow from the mining areas, while the mine tailings should undergo environmental remediation practices. It is also necessary that all wastewater, originating from the neighboring urban sites, undergo treatment before being discharged in the lagoon.

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