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# Effects of 12 years' operation of a sewage treatment plant on trace metal occurrence within a Mediterranean commercial sponge (*Spongia officinalis*, Demospongiae)

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#### Abstract

The present field study uses *Spongia officinalis* for assessing trace metals occurrence in time and space within Mediterranean rocky communities. Nine sites were selected in the Marseille area for studying spatial trends in 12 metal concentrations. Long term changes in 8 metal concentrations were assessed at sites that had been sampled before and 12 years after the opening of a treatment plant. *Spongia officinalis* highly concentrated all the trace metal surveyed excepted Hg and Cd. The overall contamination level registered provided a classification of the study sites which is congruent with that given by other studies on pollutant accumulation in neighbouring sandy-bottoms or benthic assemblages. Among the metals studied, Fe, Pb, Cr are those that best highlighted a pollution gradient. In the present study, only Cd concentration did not vary in space. Except for Ni, all pollutant concentrations clearly decreased between 1984 and 1999. This very impressive decrease in heavy metal concentrations within the Marseille area represents an indisputable evidence of the improvement of the seawater quality resulting from 12 years' operation of the Marseille sewage plant. Moreover, the significant decrease also recorded in the reference population at Port-Cros might reflect an overall improvement in the seawater quality of the NW Mediterranean.

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#### 1. Introduction

Sponges have some of the characteristics of good bioindicators and are convenient tools for characterising the state of a marine ecosystem (Battershill and Abraham, 1999; Perez, 2000). Sponge communities or indicator sponge species have frequently been used to reveal environmental crises in temperate and in tropical ecosystems (Muricy, 1991; Carballo et al., 1996; Holmes, 1996). Because of their feeding habits, with no other selection of the nutrient particles than the size of the inhalant pores, these actively pumping organisms can be considered as "biological particle traps". Sponges can thus collect and concentrate a wide range of pollutants from both the suspension and dissolved phases. High concentrations of pollutants have been reported in several species: hydrocarbons (Féral et al., 1979; Zahn et al., 1981), organochlorinated compounds (Verdenal et al., 1990; Arnoux et al., 1992; Perez et al., 2003), and metals (Patel et al., 1985; Verdenal et al., 1990; Richelle-Maurer et al., 1994; Hansen et al., 1995; Webster et al., 2002; Cebrian et al., 2003; Perez et al., 2004). Experimental studies have shown that accumulation is a function of the quantity present in the environment

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and that bioconcentration factors may be very high (Richelle-Maurer et al., 1994; Hansen et al., 1995; Cebrian et al., 2003; Perez et al., 2003). However, in spite of proposals for sponge-watch programs (Patel et al., 1985; Hansen et al., 1995), these organisms have surprisingly been little used for field surveys of contamination levels.

Spongia officinalis, one of the commercial sponges, is a Mediterranean sessile filter feeder, which is widely distributed in the various basins, mainly on rocky bottoms from 1 to 100 m depth (Mediterranean infra and circalittoral). In addition this species is characteristic of several communities, which are hotspots of biodiversity (coralligenous cliffs, overhangs, and caves). Its aquiferous system is sophisticated and able to actively filter a water volume equivalent to its own volume within a few seconds. Spongia officinalis has some of the fundamental characteristics of the ideal biomonitor proposed by Rainbow and Phillips (1993). This species is not strictly speaking opportunistic, but it is tolerant to physicalchemical fluctuations, being thus abundant at both polluted and clean sites. It is easy to collect and identify underwater, and individuals have the ability to regenerate the cutting parts within a few months. Several studies have already been conducted on this species, showing its ability to strongly accumulate metals in higher concentrations than those found in other biomonitor species, to reflect accurately the level of contamination at a given site (Verdenal et al., 1990; Perez et al., 2004) and to degrade some organic compounds (Perez et al., 2002a; Perez et al., 2003).

In the vicinity of the city of Marseille, the area known as Cortiou Cove (Fig. 1) is strongly influenced by sewage from Marseille and its suburbs, and is one of the most polluted places in the French Mediterranean. Of the whole of the Mediterranean Sea, this is also the place where the effects of sewage on both the environment and the biota are best known (for a review see Bellan et al., 1999). A daily output of about 250,000 m<sup>3</sup> is discharged into the sea, the sewage flow being mainly carried westwards by the currents and prevailing winds (Fig. 1). This transport process results in the dilution of the polluted waters, the deposition of contaminants on the sea floor and the establishment of a pollution gradient in the Cortiou Cove. The impact on macrobenthic communities of this contaminated deposit along the pollution gradient has been recorded since 1965 in a number of studies (for a review, see Bellan et al., 1999). The Marseille sewage was untreated until 1987 and has since that date been treated by a primary plant (chemically assisted sedimentation). The treatment process is at present wellsuited for filtering out suspended material (>2  $\mu$ m) and is also effective for some contaminants (e.g. from 40% for copper to 75% for lead; sewage plant monitoring unit, pers. comm.). Only iron is still discharged in excessive quantities, because of the use for the treatment process of around 5 tonnes per day of Cl<sub>2</sub>Fe. A secondary treatment process (biological) is currently at the planning stage. These measures will be followed soon by the implementation of chemical and ecological surveys of the area in accordance with national and European regulations (Directive CE 2000/60/EC).

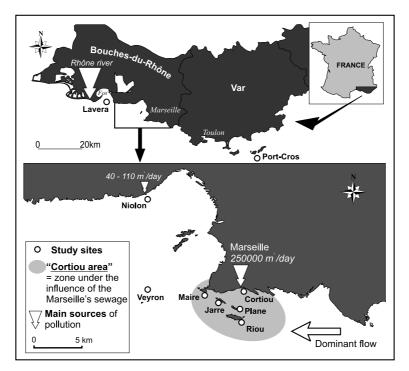


Fig. 1. Location of study sites and main sources of contamination off Marseille and within the Cortiou area.

Although they are key components of Mediterranean marine biodiversity, the state of health of deeper rocky sub-littoral communities is not regularly surveyed. The present field study uses S. officinalis for biomonitoring trace metals occurrence within these communities. Verdenal et al. (1990) analysed eight metal concentrations in this species collected in the Cortiou area 3 years before the opening of the present sewage plant. Fifteen years later, we performed an analysis of twelve metals in the same sites and three additional localities in order to assess spatial trends in an extended zone and long term changes in metal concentrations within sponges of the Cortiou area. We thus provide baselines for further surveys to be implemented in the area after the opening of the secondary treatment plant in the near future. With S. officinalis, we provide data for the French coastal quality assessment system (SEQ) under development in accordance with the new European Water Framework Directive (Henocque and Andral, 2003).

#### 2. Material and methods

#### 2.1. The species

The commercial sponge *S. officinalis* Linneus, 1759 belongs to the Dictyoceratida order. *Spongia officinalis* has a well-developed organic skeleton of spongin fibres, which include a variable amount of sand and other foreign material. However, the mineral fraction does not exceed 20% of the overall mass of the organism. In view of the large number of microbial symbionts that it hosts, this species may be considered as a "bacteriosponge" (Reiswig, 1981; Vacelet and Donadey, 1977).

#### 2.2. Study sites and sampling

Spatial trends in metal concentrations were studied at 9 sites. Five sites (Cortiou, Maire, Jarre, Plane, Riou) are located in the Cortiou area near Marseille (Fig. 1) under the more or less intense influence of the Marseille sewage. The site Cortiou, located 300m east of the discharge outlet, is heavily polluted. Maire is considered as a moderately polluted site, whereas Jarre, Plane and Riou are sites with sub-normal conditions. Two additional polluted sites were selected outside the Cortiou area. Niolon is located a few meters from a small sewer outlet and sponges there are strongly influenced by light domestic effluent. The Lavera sampling site is located in the vicinity of an industrial complex and near the mouth of the Rhone river. The other sites are located in feebly perturbed areas. The Le Veyron sampling site is 10km off Marseille and could be considered as a clean site. Port-Cros was the reference site of the study conducted by Verdenal et al. (1990). Located in a National Park, 30km off the Provence Coast, 100km from Marseille,

this site is characterised by a strong hydrodynamism only exposed to the northwestern current. Among these 9 sites, long term changes were assessed at 6 sites (Cortiou, Maire, Jarre, Plane, Riou, Port-Cros) that had been sampled before (Verdenal et al., 1990) and 12 years after the opening of the treatment plant. For studying the spatial variations in metal concentrations, six specimens per sampling site were collected by scuba diving in summer 1999, i.e. during the same season as sampling by Verdenal et al. (1990) in 1984. The specimens were collected at a depth of between 15 and 20m on vertical rock-faces or on the ceiling of small caves, where they are protected from sedimentation. Specimens of similar size were chosen in order to avoid as far as possible variations of metal concentrations due to differences in age.

#### 2.3. Treatment of the samples for metal analysis

The samples were frozen at -25 °C soon after collection, and subsequently lyophilised. One gram of each freeze-dried sample was mineralized with 10ml of nitric acid 65% Merck (suprapur quality) in PTFE bombs using a microwave oven (Prolabo A301 Microdigest). The digests were analyzed for 12 metal elements: manganese, nickel, iron, copper and zinc by flame atomic absorption spectrometry (PYE UNICAM SP9 Atomic Absorption Spectrophotometer Philips); lead and cadmium by a graphite furnace atomic absorption spectrometry (UNICAM 939 QZ Atomic Spectrophotometer Absorption with graphite furnace "Zeeman 6790" SOLAR); chromium, arsenic, titanium and vanadium by Inductively Coupled Plasma Spectrometry (JOBIN YVON "JY24"); and mercury by X-ray Fluorescence (Merlin Mercury Fluorescent Detector). The analytical methods were validated through internal and external calibration (Quality Assurance and Quality Control approved by the French Ministry of the Environment). The results given as means and standard deviations expressed in µg/g of dry weight were analyzed statistically for differences between the sites by the Kruskall-Wallis test. Student T tests were applied for differences between concentrations recorded in 1999 and 1984. Cluster analvsis of the global metal content of the sponges provided classifications of the study sites before and after the opening of the sewage plant. All statistical methods were applied using the software STATISTICA 5.1 (Statsoft France).

#### 3. Results

#### 3.1. Spatial trends

The mean concentrations and standard deviations of the 12 trace metals measured at 9 sites in 1999 are detailed in Table 1.

|           |   | Fe      | Mn    | Ni   | Cu   | Zn   | Pb   | Cd   | As    | Cr   | V    | Ti   | Hg  |
|-----------|---|---------|-------|------|------|------|------|------|-------|------|------|------|-----|
| Cortiou   | т | 5823.7  | 31.6  | 45.9 | 50.5 | 58.8 | 9.3  | 0.4  | 98.8  | 9.8  | 61.9 | 37.0 | 0.8 |
|           | S | 1014.6  | 22.3  | 5.7  | 5.9  | 3.2  | 3.1  | 0.03 | 11.9  | 0.9  | 1.5  | 2.6  | 0.1 |
| Maire     | т | 2155.6  | 17.2  | 50.5 | 37.7 | 59.0 | 1.8  | 0.3  | 86.3  | 6.7  | 49.5 | 26.2 | 0.5 |
|           | S | 572.2   | 1.9   | 5.5  | 2.8  | 10.1 | 0.4  | 0.03 | 1.9   | 0.9  | 2.2  | 1.4  | 0.1 |
| Plane     | т | 321.3   | 14.3  | 51.6 | 41.6 | 48.5 | 0.6  | 0.3  | 104.6 | 4.9  | 44.9 | 21.9 | 0.9 |
|           | S | 40.3    | 0.6   | 5.0  | 6.7  | 5.7  | 0.2  | 0.1  | 13.5  | 0.3  | 2.6  | 1.1  | 0.3 |
| Jarre     | т | 827.9   | 41.3  | 52.5 | 39.6 | 47.3 | 1.8  | 0.4  | 93.5  | 5.9  | 47.6 | 23.6 | 0.6 |
|           | S | 253.3   | 21.1  | 7.4  | 5.6  | 4.2  | 0.5  | 0.1  | 2.5   | 0.8  | 1.5  | 1.5  | 0.1 |
| Riou      | т | 343.5   | 24.2  | 49.3 | 41.3 | 46.1 | 0.6  | 0.4  | 119.9 | 5.3  | 42.0 | 23.0 | 0.5 |
|           | S | 161.0   | 5.0   | 2.9  | 10.0 | 2.0  | 0.1  | 0.1  | 4.4   | 0.9  | 3.5  | 1.7  | 0.1 |
| Port-Cros | т | 363.4   | 10.9  | 46.8 | 36.6 | 46.3 | 0.8  | 0.3  | 113.0 | 4.8  | 43.8 | 22.2 | 0.6 |
|           | S | 203.5   | 2.0   | 5.7  | 7.1  | 1.8  | 0.4  | 0.1  | 8.4   | 0.2  | 1.4  | 0.9  | 0.4 |
| Veyron    | т | 455.0   | 20.8  | 54.5 | 33.3 | 47.8 | 0.7  | 0.5  | 120.7 | 5.0  | 44.8 | 23.4 | 0.6 |
|           | S | 25.4    | 1.3   | 3.2  | 0.3  | 1.0  | 0.1  | 0.2  | 2.3   | 0.3  | 1.0  | 0.8  | 0.1 |
| Lavera    | т | 8820.5  | 182.4 | 60.6 | 48.9 | 80.3 | 7.0  | 0.3  | 134.1 | 7.5  | 76.9 | 33.7 | 0.5 |
|           | S | 285.1   | 28.8  | 1.6  | 0.9  | 3.3  | 0.2  | 0.1  | 1.5   | 0.1  | 1.1  | 1.1  | 0.1 |
| Niolon    | т | 16258.4 | 15.2  | 42.8 | 71.3 | 79.2 | 15.6 | 0.3  | 152.4 | 10.1 | 58.2 | 19.0 | 1.1 |
|           | S | 188.1   | 0.8   | 9.2  | 0.4  | 0.8  | 4.8  | 0.1  | 2.5   | 0.3  | 1.2  | 0.1  | 0.5 |

Table 1 Mean (*m*) and standard deviation (*s*) of heavy metal concentrations ( $\mu g/g$  of dry weight = ppm) in sponges (6 specimens per sampling) collected in summer 1999

Cluster analysis was performed to compare the overall concentrations of 12 trace metals recorded in 1999 between 9 sites (Fig. 2). Two groups of sites show approximately 60% divergence. A first cluster includes the sites of Jarre, Veyron, Riou, Plane, Maire and the reference site of Port-Cros. Within this group, overall metal concentrations appear to be similar with the exception of Maire. A second cluster includes Lavera, Cortiou and Niolon. Lavera and Cortiou have the closest metal concentrations (10% divergence) and a difference of about 30% is observed between the two sites and Niolon.

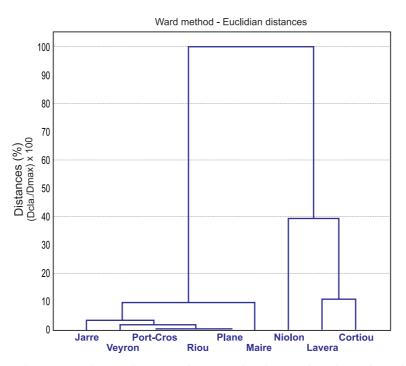


Fig. 2. Cluster analysis performed to compare the overall trace metal concentrations between the 9 sites under study in 1999 (Ward method and Euclidian distances).

*Iron.* Highly significant differences were recorded between the 9 sites (H = 38.7, df = 8, p < 0.001). The highest values were at Niolon, Lavera and Cortiou in that decreasing order (16258.4 to 5823.7 µg/gDW). For the Cortiou area, the concentration in sponges at Maire was higher than at Jarre, Plane and Riou. These differences are consistent with the overall comparison of metal levels (Fig. 2).

*Lead.* Concentrations measured at all sites showed highly significant differences (H = 38.3, df = 8, p < 0.001). The highest values were at Niolon, Cortiou and Lavera in that decreasing order (15.6 to  $7.0 \mu g/g$ DW). For the Cortiou area, sponges at Maire and Jarre showed iron concentrations that were more than twice the concentration at Plane and Riou. With the exception of Jarre, this overall variation pattern is also suggested by the cluster analysis (Fig. 2).

*Chromium.* Accumulated concentrations showed highly significant differences between all locations (H = 35.2, df = 8, p < 0.001). Maximum levels that are nearly identical were recorded in sponges at Cortiou and Niolon (respectively 9.8 and  $10.1 \mu g/g DW$ ). Concentrations at Lavera were higher than at all remaining sites. For the Cortiou area especially, concentrations at Maire were higher than at Jarre, Plane and Riou. These spatial trends are in good agreement with the cluster analysis (Fig. 2).

*Vanadium*. Variations between all sites were highly significant (H = 35.4, df = 8, p < 0.001). The highest concentration was in sponges at Lavera (76.9 µg/ gDW). Concentrations at Cortiou and Niolon were similar and higher (respectively 61.9 and 58.2 µg/gDW) than at all remaining sites. In the Cortiou area, concentrations appeared relatively similar. With the exception of Maire, this overall variation pattern is also suggested by the cluster analysis (Fig. 2).

*Copper.* Concentrations were significantly different between the 9 sites (H = 23.3, df = 8, p < 0.01). The highest concentration was recorded in sponges at Niolon (71.3 µg/g DW). Concentrations at Cortiou and Lavera were close (respectively 50.5 and 48.9 µg/g DW) and still higher than at remaining sites. Concentrations at Maire, Jarre, Plane and Riou were homogeneous (from 37.7 to 41.6 µg/g DW), the lowest concentrations being found in sites away from the coast (Le Veyron and Port-Cros). With the exception of Maire, these spatial trends are consistent with the cluster analysis (Fig. 2).

*Titanium.* Concentrations varied highly significantly among all sites (H = 38.2, df = 8, p < 0.001). The highest concentrations were at Cortiou and Lavera (respectively 37.0 and 33.7 µg/gDW), and Niolon showed the lowest concentration (19.0 µg/gDW). Values at all other stations were similar. In the Cortiou area, this distribution pattern of concentrations is in agreement with the cluster analysis (Fig. 2). *Zinc.* Concentrations also differed highly significantly between all sites (H = 32.3, df = 8, p < 0.001). The highest levels were recorded at Niolon and Lavera (79.2 and  $80.3 \mu g/g DW$ ). Concentrations in sponges at Cortiou and Maire were higher than at all remaining sites. Sponges at the other sites showed very similar concentrations. These differences are generally congruent with the overall comparison of metal levels (Fig. 2).

Arsenic. Concentrations differed highly significantly between the 9 sites (H = 36.0, df = 8, p < 0.001). The highest arsenic concentrations were also at Niolon and Lavera (respectively 152.4 and 134.1µg/gDW). For the Cortiou area, however, Plane and Riou showed the highest levels and concentrations at Maire, Cortiou and Jarre were the lowest.

*Manganese*. Highly significant differences were observed between all sites (H = 31.8, df = 8, p < 0.001). A very high concentration of manganese was recorded in sponges at Lavera (182.4µg/gDW). Values at all other sites were much lower but showed not inconsiderable variations. For the Cortiou area, the highest concentration values were at Jarre and Riou (respectively 41.3 and 24.2µg/gDW).

*Mercury.* Concentrations were rather low and the highest values were at Cortiou, Niolon and Plane (0.8–1.1 µg/g DW). Still lower but variable concentrations were recorded at all other locations. These values resulted in highly significant differences in concentrations between all sites (H = 26.6, df = 8, p < 0.001).

*Nickel.* Only slightly significant variations were recorded between all sites (H = 15.6, df = 8, p = 0.0482). Lavera showed the highest concentrations ( $60.6 \,\mu g/g$  DW) and the lowest were recorded at Niolon ( $42.8 \,\mu g/g$  DW). Relatively homogeneous nickel levels characterised other sites.

*Cadmium.* Concentrations are very low  $(0.1-0.4 \mu g/g DW)$  and variations between sites were not significantly different (H = 14.7, df = 8, p = 0.0652). The highest and lowest cadmium concentrations corresponded to sponges collected at Veyron and Port-Cros respectively.

#### 3.2. Long term changes

Trace metal concentrations were measured in *S. officinalis* during the summer of 1984 (Verdenal et al., 1990), 3 years before sewage treatment which started in 1987 at Marseille. Metal levels in sponges were measured at Cortiou, Maire, Plane, Jarre, Riou and at the reference site of Port-Cros for 8 elements (Mn, Ni, Fe, Cu, Zn, Pb, Cd, Cr). Only mean concentrations and standard deviations are available and cadmium concentration in sponges was not measured at Maire and Plane in 1984. Fig. 3 presents mean concentrations and standard deviations of those 8 metals recorded in 1984, and shows the differences with concentrations recorded

in 1999. With the exception of nickel, all metal concentrations recorded in 1999 at all sites are lower than those recorded in 1984. For copper, zinc, lead, cadmium and chromium, the decrease observed at all sites except Port-Cros is significant at  $p \leq 0.01$ . Concentrations of copper, lead, cadmium and chromium recorded at Port-Cros also decreased significantly ( $p \leq 0.05$ ) but the reduction in zinc observed at Port-Cros is not significant (p > 0.05). Differences in iron concentrations at Maire and Riou are not significant (p > 0.05), nor are the variations in manganese detected at Cortiou and Jarre (p > 0.05). Decreases in iron and in manganese recorded at Port-Cros are significant ( $p \leq 0.05$ ). Nickel showed a reverse trend and a significant increase in concentration was recorded at Maire, Plane and Riou  $(p \leq 0.01)$ . Nickel concentrations recorded at Port-Cros, Jarre and Cortiou in 1984 and in 1999 did not differ significantly (p > 0.05).

We performed a cluster analysis to compare the overall trace metal concentrations observed in S. officinalis collected at 6 sites in 1984 and in 1999. Each site is therefore represented twice in Fig. 4. Two clusters of measures diverge by 60%. A first cluster includes concentrations recorded at Cortiou in 1984 and in 1999, and the two measures differ by approximately 30%. A second cluster includes all other concentrations and is composed of two subgroupings diverging by about 30%. A first sub-group is composed of overall metal concentrations recorded at Maire in 1984 and in 1999, and values observed at Jarre and Plane in 1984. A second sub-group is defined by global metal concentrations at the reference site Port-Cros and at Riou in 1984 and in 1999. This sub-group also includes concentrations recorded at Jarre and Plane in 1999. Therefore, changes of about 30% in overall metal concentrations occurred at Jarre and Plane between 1984 and 1999 (Fig. 4).

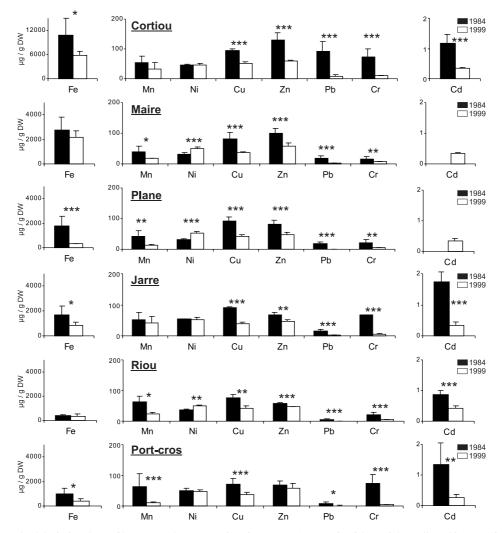


Fig. 3. Mean and standard deviation (bar) of heavy metal concentrations in sponges ( $n = 6 \mu g/g$  of dry weight) collected in 1984 (from Verdenal et al., 1990) and 1999 at the six common study sites. Results of the Student *T* tests on concentrations are given by the significance levels of the differences as follows: \*p < 0.05; \*\*p < 0.01; \*\*\*p < 0.001.

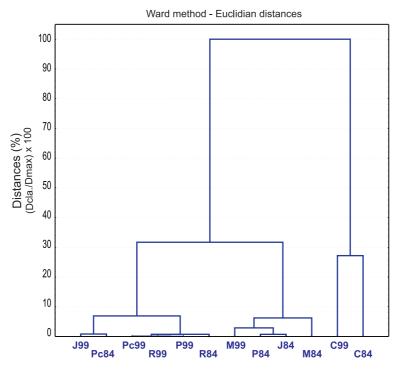


Fig. 4. Cluster analysis performed to compare the overall trace metal concentrations between the 6 common study sites in 1984 and 1999. Sites: C = Cortiou; M = Maire; J = Jarre; P = Plane; R = Riou; PC = Port-Cros (Ward method and Euclidian distances).

#### 4. Discussion and conclusion

A single biomonitor gives only a partial image of pollutant bioavailability, which depends notably on its habitat and its life strategy. That is the reason why the use of a range of species representative of various feeding strategies or trophic levels and of different habitats is now considered essential to obtain an overall view of pollutant bioavailability in coastal ecosystems (Rainbow, 1995). According to Rainbow and Phillips (1993), a complete set of biomonitors might include organisms able to accumulate dissolved sources of pollutant and/or to uptake particles of variable sizes. In this study, sponges have been chosen among other sessile invertebrates dwelling on rocks because of their life strategy and unmatched potential for responding to environmental fluctuations (Perez, 2000).

Spongia officinalis highly concentrate all the trace metal surveyed excepted Hg and Cd. The overall contamination level registered in *S. officinalis* provided a classification of the study sites which is consistent with knowledge of pollutant dispersion in the Cortiou area and their accumulation in sediments (Bellan et al., 1999; Arfi et al., 2000). This classification is also congruent with that provided by the ecological data on sponges (Muricy, 1991) and other assemblages (Harmelin and Capo, 2002; Perez et al., 2002b). Spatial variations in metal bioavailability is in fairly good agreement with results from previous works on *S. officinalis* as biomonitor in the same area (Verdenal et al., 1990; Perez et al., 2003). Among the metals studied, Fe, Pb, Cr are those that best highlighted a pollution gradient resulting from the urban sewage of Marseille. Probably mainly spread with coarse particles, these elements are thus good indicators of the Marseille sewage's influence within the Cortiou area. Verdenal et al. did not find any significant spatial fluctuations of Cu, Mn and Cd concentrations in sponges. In the present study, only Cd concentration did not vary in space. Our results are also consistent with those of Verdenal for Mn concentration, which did not vary significantly within the Cortiou area, and was only considerably higher in sponges collected at Lavera, near the Rhône estuary. Mn could be thus confirmed as a good indicator of the Rhône river influence (Malard et al., 1998; Younes, 2000). Cu concentration was also relatively homogenous within the area, except in the sites nearest to a source of contamination: urban source at the Cortiou and Niolon sites and industrial source in Lavera. This pattern indicates that Cu has multiple origins within the Marseille area and is mainly spread with fine particles.

The objectives of the treatment process implemented in 1987 were first to filter out the largest and heaviest particles, to regulate the effluent flow and to reduce as far as possible the spread of fine particles. The retention of suspended material led to a significant reduction of pollutant inputs within the Cortiou area. This study confirms that *S. officinalis* as a biomonitor allows detection of temporal changes in pollutant bioavailability (Perez et al., 2003). In spite of the processing of the sewage, the sponge population at Cortiou still appeared in 1999 to be the most contaminated one. The contamination status of Maire was moderate in 1999, but there, the contamination level of the sponge population was relatively similar to those of the Maire, Jarre and Plane populations before the opening of the sewage treatment plant (Fig. 3). On the other hand, the contamination levels of two sponge populations, Plane and Jarre, have been considerably reduced. Although they were considered in 1984 as moderately polluted, in 1999 their pollution level is closest to the cleanest population at Port-Cros (Fig. 3).

Except for Ni, all pollutant concentrations clearly decreased between 1984 and 1999. The differences in Fe concentration were not significant for the Maire and Riou sites near Marseille. The huge amount of Fe still recorded in sponges from the Marseille area could be explained by the use of ferric chloride as an agent of coagulation for the physical-chemical treatment of the sewage. The most spectacular decrease, interpreted as a consequence of the filtering out of the largest particles (>2 $\mu$ m), was observed for Cr and Pb. The recording of Pb concentrations 10 times lower than in 1984 is also probably also due to the increasing use of lead-free fuel since the beginning of the 1990s. Besides, a drastic decrease in PCB concentrations in sponges has also been recorded at Cortiou, from 17 µg/g in 1984 to an average of 4µg/g in 1999 (Perez et al., 2003).

According to data obtained by the National Marine Environment Monitoring Network (RNO), the decrease in heavy metal contamination in the Cortiou area is not so clear and depends on the compartment studied. The comparison of data collected over 20 years is sometimes contradictory. In seawater, after the beginning of sewage treatment, the occurrence of low pollutant concentrations in seawater decreased in the vicinity of the sewage whereas episodes of high levels of contamination became less frequent (RNO QUADRIDGE database). Mussels better recorded the consequences of the improvement of the seawater quality than did sediments. In 1993, whereas the improvement of quality of sediments was barely marked, the concentrations in mussels from Cortiou were already 30% lower for Hg, Pb and Zn and 75% lower for PCB (Arnoux et al., 1993). These observations are fully consistent with data obtained from sponges in 1999. But the results obtained from mussels under a weaker influence of the sewage are quite different. For example, Cd and Zn concentrations did not significantly decrease in mussels. Concentrations of these two elements have fluctuated a lot, and the mean concentrations for the 80s and the 90s are not significantly different  $(163.5 \pm 31 \,\mu\text{g/g})$  versus  $175.4 \pm 25 \,\mu\text{g/g}$ for Zn;  $1.1 \pm 0.4 \,\mu\text{g/g}$  versus  $1.0 \pm 0.2 \,\mu\text{g/g}$  for Cd). Fluctuations of Hg, Cu and Pb were less pronounced during the 90s. The difference between the mean concentrations of the two periods is not significant for Hg, but it is

for Cu  $(7.0 \pm 1.5 \,\mu\text{g/g} \text{ versus } 5.6 \pm 0.7 \,\mu\text{g/g})$  and Pb  $(5.1 \pm 2.0 \text{ versus } 3.2 \pm 0.5 \,\mu\text{g/g})$  (RNO QUADRIDGE database).

Despite these mixed results concerning the changes in the seawater quality off Marseille, important shifts in the composition of marine communities in the Cortiou area have been attributed to the sewage treatment. The recovery of the benthic assemblages has been faster than expected (Bellan and Bourcier, 1993). Recovery is most noticeable within the polluted zone and seems slower in sites less influenced by the effluent (Bellan et al., 1999). Less conspicuous changes in macroalgal communities have also been observed (Soltan et al., 2001): the Cystoseira amantacea community, well known as an indicator of clean waters (Bellan-Santini, 1968; Belsher, 1979), has particularly to be restored. Thus, the improvement of the quality of the marine environment is most clearly apparent in sites in the vicinity of the sewage. It is generally more pronounced in biotic than in abiotic compartments, and the recovery of the communities seems to be faster than the decrease in contamination levels. In a zone further from a strong anthropic disturbance, but obviously influenced by different distant sources of contamination, the decrease in pollutant concentrations is less pronounced in mussels than in sponges.

The very impressive decrease in trace metal concentrations within sponges from the Cortiou area under the strong influence of a sewage outlet represents indisputable evidence of the improvement of the seawater quality resulting from 12 years' operation of the Marseille sewage plant. Moreover a significant decrease has also been recorded in the reference population at Port-Cros. Despite the lack of chemical baselines in those pristine marine communities, the observed decrease might reflect an overall improvement in the seawater quality of the NW Mediterranean.

This study is one of the first field applications of sponges as biomonitor and clearly demonstrates that these organisms living in circalittoral rocky communities are well suited for providing an overall view of contaminant bioavailability. They may thus provide additional information to that provided by usual biomonitors at other depth ranges or within different marine communities and are well suited for large-scale and long term monitoring programmes in the Mediterranean Sea.

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