Assessment of the Environmental Significance of Heavy Metals Pollution in Surficial Sediments of South Brittany Waters, France: An Ecological Risk Assessment Approach

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48

49 **Abstract**

50 Distribution patterns of selected heavy metals content in sediments from the Bay of Quiberon

51 and Gulf of Morbihan were studied to understand the current heavy metals contamination due

52 to urbanization and mariculture activities in the coastal area. Therefore, a survey was

53 conducted and 196 sediments collected were characterized for heavy metals content using

54 Inductively Coupled Plasma – Mass Spectrometry (ICP–MS) after mix acid digestion process.

55 The distribution maps of the concentrations of the heavy metals studied were produced as an

isopleth map based on data interpolation by the ArcGIS software application. The association with the adverse effects on aquatic organisms was determined by the classification of the sediment according to the sediment quality guidelines. Therefore, two approaches were employed namely; direct comparison with Sediment Quality Guidelines (SQGs) by USEPA and comparison with other numerical SQGs, threshold effect level/probable effect level, and effect range low /effect range medium. In order to estimate the effect of multiple contaminations of heavy metals, the mean-ERM-quotient was calculated at each sampling point.

Keywords

Heavy metals, Bay of Quiberon, Gulf of Morbihan, sediment, sediment quality guidelines

Introduction

Nowadays, estuary and coastal environments are contaminated by human activities containing elevated concentrations of heavy metals, nutrients, organic pollutants, and radionuclides (Clark et al. 2000; Kennish 2002). Some chemicals are highly toxic and persistent and have a strong tendency to concentrate in marine food webs. The pollution of coastal zones near metropolitan areas, caused by these anthropogenic wastes is due to the large human

74 population and the enormous amounts of sewage discharged into coastal waters (Sadiq 2002;
75 Bay et al. 2003). The addition of waste products into rivers and estuaries, particularly those in
76 industrial and population centres has led to a significant increase in contamination of heavy
77 metals (Jayaprakash et al. 2005). Accumulation of heavy metals in surface sediments from
78 industrial effluents and urban discharged into the river without proper cleaning is easily
79 identified by heavy metals spatial variations in sediments (Dauvalter & Rognerud 2001).

80

81 Coastal areas are sensitive to both climate changes and human impacts, which make these
82 areas ideal for studying environmental changes and the sequence of coastal sediments, and
83 have become attractive targets for documenting paleoenvironmental changes. The Bay of
84 Quiberon and Gulf of Morbihan are an excellent example of the coastal sea with high
85 terrestrial influence, a semi-enclosed bay, surrounded by densely populated areas and
86 mariculture activities. Such an investigation is required if the environmental impacts of the
87 region are to be understood quantitatively and qualitatively.

88

89 In this exploration, surficial spatial distributions of selected heavy metals; Copper (Cu), Zinc
90 (Zn), Cadmium (Cd), Lead (Pb) (identified as priority contaminants in aquatic systems by the
91 European Community (EC) Water Framework Directive 2000/60/EC; Tessier et al. 2011)

were characterized in the surficial sediments which reflects the recent input to evaluate the contamination level resulting from anthropogenic inputs. This EC work recommends the monitoring of fluvial loads of heavy metals as a tool for sustainable management in the aquatic ecosystem (Masson et al. 2006).

Metals are toxic because they bioaccumulate in which the accumulate in organisms and are higher than in the environment (Gupta 2013; Goretti et al. 2016; Sobihah et al. 2018; Tengku Nur Alia et al. 2020). Furthermore, metals also biomagnify, amplifies through the food chain and accumulates in the top predators (Storelli et al. 2005; Gu et al. 2017; Rodríguez-Jorquera et al. 2017; Bonsignore et al. 2018). Trace metals are also very persistent and difficult to be removed once they enter the body of the organisms (Storelli et al. 2011; Benali et al. 2017; Gu et al. 2018).

Aim of Research

The current study aims to analyze the levels of four heavy metals (Cu, Cd, Pb and Zn) at 196 sampling points in the Bay of Quiberon and Gulf of Morbihan surficial sediments. The adverse effects on aquatic organisms were determined by direct comparison with the Sediment Quality Guidelines (SQGs) and the sediment toxicity and ecological risk indices,

namely the effect low range (ERL) / effect range median (ERM), and the threshold effect level (TEL) / probable effect level (PEL) values. In order to estimate the effect of multiple contaminations of heavy metals in the study area, the mean-ERM-quotient was also calculated at each sampling point.

Methods/Experimental

Study Area

Bay of Quiberon is a large open-sea area on the south coast of Brittany, consisting of two additional areas with an equivalent surface area of approximately 120 km² (Figure 1). The bay is roughly triangular, almost completely surrounded by land on the north and east sides; open to the south with the Gulf of Morbihan to the north-east and the narrow peninsula of Quiberon providing protection from the Atlantic Ocean to the west. The Belle, Houat, and Hoëdic islands add to the protection of the bay (Proust et al. 2001; Chaumillon et al. 2008).

Gulf of Morbihan (“Mor-bihan” = “little sea” in Breton) is a natural harbor located in the South Brittany, northwest France (Figure 1). It is a close sea body (11 500 ha) and a shoreline of 250 km (Menier et al. 2011). It is a shallow basin with a maximum depth of 30 m and is connected to the sea by a 900 m wide and 30 m deep tidal inlet at Port Novalo (Rossi et al.

2011). Institut Géographique National France recorded that there are 37 islands located in the gulf where the two largest islands are Moine and Arz Islands. The gulf is sheltered from the Atlantic Ocean (Bay of Biscay) by the peninsula of Quiberon, Houat, Hoëdic, and Belle Islands.

Many areas along the coast are used by the public as recreation areas. A number of shipping ports for recreational purposes and fishing activities are located in this region. In addition, the surrounding coast is successfully used for commercial shellfish (oyster and mussel) mariculture activities with a significant market around the region.

Sample Collection

This oceanographic fieldwork was carried out by the SEPIOLA research vessel of the University of Rennes1 at the Gulf of Morbihan and vessel of the Regional Committee of Conchyliculture (CRC), South Brittany at the Bay of Quiberon. A total of 196 surficial sediments were collected randomly in Bay of Quiberon and Gulf of Morbihan with Orange Peel Grab sampler. In addition, in order to prevent metals contamination from the wall of the grab, the outermost layer of the sediment samples were removed and only the inner part was kept for further analysis. After collected, the samples were stored in plastic containers and

kept at low temperatures until analysis was carried out.

Laboratory Analysis

Sediment samples were digested and analyses for total heavy metals following published methodologies with some modifications (Ong & Kamaruzzaman 2009; Kamaruzzaman et al. 2008; Kamaruzzaman et al. 2010). The digestion method involved heating of 50mg of a finely powdered sample in a sealed Teflon vessel in a mixture with a mixed acid solution of concentrated HF, HNO₃ and HCl. The Teflon vessel was kept in an oven at 150°C for 6 hours. After cooling at room temperature, the solution of the vessel was transferred into a polypropylene test tube. An inductively coupled plasma mass spectrometer, ICP-MS (Perkin Elmer Elan 9000) was used for the quick and precise determination of selected heavy metals in the digested sediment samples. The accuracy was also examined by analyzing duplicate Standard Reference Material 1646a Estuarine Sediment, the results of which were within $\pm 3\%$ of certified values.

Results and Discussion

All the analyzed data were visualized using ArcGIS software and presented in the form of a concentration isopleth map to identify the sediment concentration hotspot as shown in Figure

2. Visualization by overlaying maps using a geographical information system makes this presentation even easier and more successful (Caeiro et al. 2005).

Tomlison et al. (1980) elaborated that the application of the pollution load index (PLI) provides a simple way to assess the quality of estuarine and coastal sediment. This assessment is a quick tool for comparing the status of pollution at different locations (Karbassi et al. 2006). PLI represents the number of times the concentration of heavy metals in the sediment exceeds the background concentration and provides a summary indication of the overall level of heavy metal toxicity in a particular sample or location (Rabee et al. 2011). The PLI can provide the public in the surrounding area with some understanding of the quality of the components in their environment, indicating the trend both spatially and temporarily (Harikumar & Jisha, 2010). In addition, it provides valuable information to decision-makers towards better management of pollution levels in the Bay of Quiberon and Gulf of Morbihan regions.

PLI is obtained as contamination factors (CF). This CF is the quotient obtained by dividing the concentration of each metal with the background value of the metal proposed by Wedepohl (1995). The PLI can be expressed in the following calculation:

182
$$\text{Pollution load index (PLI)} = (CF_1 \times CF_2 \times CF_3 \times CF_n)^{1/n}$$

183 where, n is the number of metals (4 in the present study) and CF is the contamination factor.

184 Background value : Cu 55 µg/g; Cd 0.2 µg/g; Pb 20 µg/g; Zn 70 µg/g

185 The CF can be calculated from:

$$\text{Contamination factor (CF)} = \frac{\text{Element concentration in the sediment}}{\text{Background value of the element}}$$

186 The background value (of the elements were adopted from Wedepohl (1995) to calculate the
187 CF. The PLI value > 1 is categorized as polluted, while <1 indicates no pollution at the
188 sampling point (Chakravarty & Patgiri 2009; Seshan et al. 2010).

189

190 PLI values of the analyzed surficial sediments were visualized by ArcGIS software and shown
191 in Figure 3, which confirms that either Bay of Quiberon or Gulf of Morbihan is not seriously
192 polluted with the heavy metals studied. PLI was found to be generally low (<1) in all the
193 studied locations in the Bay of Quiberon, apart from Trinité-sur-Mer area. The eastern zone
194 and rivers (Auray, Marle and Noyal) zones also have a PLI value above 1; therefore these
195 areas can be classified as moderate contamination factors (Seshan et al. 2010).

196

197 Two approaches were used to assess sediment toxicity and ecological risk to benthic
198 organisms for heavy metals studied in Bay of Quiberon and Gulf of Morbihan surficial

sediments. The first approach was to compare the concentrations of heavy metals to the Sediment Quality Guidelines (SQGs) promulgated by the United States Environmental Protection Agency (USEPA) which were reviewed by Giesy et al. (1990). This approach was used only to screen contaminants of concern in aquatic ecosystems. On the other hand, the second approach was to apply two sets of SQGs developed for marine and estuarine ecosystems (Long & McDonald 1998) to the assessment the ecotoxicology of the concentration of heavy metals in sediments, namely, (a) the effect range low (ERL)/effect range median (ERM) and (b) the threshold effect level (TEL)/probable effect level (PEL).

Based on the SQGs proposed by USEPA from Giesy et al. (1990), sediment contamination is defined as three classes, non-polluted, moderately polluted and heavily polluted (Table 1). This classification of SQGs does consider the natural background or multiple elements (Luo et al. 2010). By comparing the mean concentrations of heavy metals in Bay of Quiberon and Gulf of Morbihan surficial sediments with SQGs, sediments in both areas were classified as non-polluted for Zn, and Pb, and moderately polluted by Cu and Cd where these metals exceed SQGs. Surficial sediments from Trinité-sur-Mer zone in Bay of Quiberon, Auray, Marle, Noyalo river zones, and the eastern in Gulf of Morbihan catchment area were moderately enriched with these metals. However, no surficial sediments from both sampling

217 areas were classified as heavily polluted with the heavy metals studied according to SQGs
218 classification.

219

220 The concentration of heavy metals were evaluated in a screening-level ecological risk
221 assessment by comparing them to another numerical SQGs, namely the threshold effect level
222 (TEL)/probable effect level (PEL), and the effect range low (ERL)/effect range medium
223 (ERM) which were proposed by Long and McDonald (1998). Low range values (i.e., ERL
224 and TEL) are less than expected to have adverse effects on sediment dwelling fauna. In
225 contrast, ERM and PEL represent chemical concentrations above which adverse effects are
226 likely to occur in the aquatic ecosystem (Long & MacDonald 1998).

227

228 The SQG values for heavy metals studied in the surficial sediments from the Bay of Quiberon
229 and Gulf of Morbihan and the sediment classification based on these guidelines are shown in
230 Table 2. Results of SQGs classification suggest that all heavy metals in sediments would
231 rarely be expected to cause adverse effects on biota that living in the area.

232

233 The majority of heavy metals studied did not exceed the corresponding ERL and TEL
234 guideline values and therefore do not pose a risk to marine organisms living in Bay of

Quiberon and Gulf of Morbihan. Only a small percentage of surficial sediments would be classified as possibly posing an occasional threat to the organism due to concentrations of Cu. However, Cr, Cu, and Pb in a certain location exceeded the TEL value, particularly from the Gulf of Morbihan surficial sediments. Thus, at a concentration greater than the TEL, the toxic effect of long-term exposure to these heavy metals would be predicted to occur in the station sampled.

In order to estimate the effect of multiple contaminations on heavy metals, mean-ERM-quotient (M-ERM-Q) was calculated for each surficial sediment in the Bay of Quiberon and Gulf of Morbihan according to Long et al. (1998a), Larrose et al. (2010) and Tessier et al. (2011) as follows:

$$M - ERM - Q = \frac{\sum_{i=1}^n (C_i / ERM_i)}{n}$$

Where C_i is the surficial sediment concentrations of element (i), ERM_i is the ERM for element (i) and n is the number of elements, e.g. four heavy metals in this study.

Long et al. (1998b) have defined several classes of toxicity probability for biota organisms in their study from US coast based on amphipod survival test: $m-ERM-Q < 0.1$ has a 9%

probability of toxicity); $0.1 < m-ERM-Q < 0.5$ has a 21% probability of being toxic; $0.5 < m-ERM-Q < 1.5$ has a 49% of being toxic; and $m-ERM-Q > 1.5$ has a 76% probability of toxicity. Using the concentrations of the heavy metals studied, the $m-ERM-Q$ value was calculated, and the obtained distribution visualized by ArcGIS software in Bay of Quiberon and Gulf of Morbihan is shown in Figure 4.

The $m-ERM-Q$ distribution pattern in the Bay of Quiberon surficial sediments showed that the inner bay zone had values below 0.1, which indicated a 9% probability of being toxic to the benthic organisms in that area. In the outer bay, the $m-ERM-Q$ values slightly exceed 0.1, but not more than 0.2 indicate that this zone is between 9% and 21% probability to be toxic to benthic organisms. However, in Trinité-sur-Mer area, $m-ERM-Q$ ranged from 0.2 and 0.3; therefore, this area can be categorized as 21% probability of toxicity to the benthic organisms according to Long et al. (1998b) classification.

Meanwhile, in the gulf of Morbihan sediments, the $m-ERM-Q$ values at the western zone of the gulf were lower than 0.1m, and therefore this zone can be categorized as 9% probability of being toxic to the benthic organisms. Towards the inner gulf of eastern zone, the values of $m-ERM-Q$ were increased but do not exceed the 21% probability of toxicity to the benthic

organisms, 0.2. Generally, some sampling locations located at the rivers zone (Auray, Marle and Noyalo) had the m-ERM-Q value higher than 0.3, which meant a 21% probability of toxicity to the benthic organisms. The impact of contaminated sediment probably on aquatic ecosystems is likely to be due to re-suspension and remobilization processes (Tankere-Muller et al. 2007; Kalnejais et al. 2010)

Abbreviations

Cd: Cadmium; CF: contamination factors; CRC: Regional Committee of Conchyliculture; Cu: Copper; EC: European Community; ERL: effect low range; ERM: effect range median; HCl : Hydrochloric acid; HF: Hydroflouric acid; HNO₃: Nitric acid; ICP-MS: Inductively Coupled Plasma – Mass Spectrometry, M-ERM-Q: mean-ERM-quotient; Pb: Lead; PEL: probable effect level; PLI: pollution load index; SQGs: Sediment Quality Guidelines; TEL: threshold effect level; USEPA: United States Environmental Protection Agency; Zn: Zinc

Declarations

Availability of data and material

Data sharing not applicable to this article as no datasets were generated or analyzed during the current study. Please contact author for data requests.

289

290 **Competing interests**

291 The authors declare that they have no competing interest.

292

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297

298 **Authors' contributions**

299 Meng-Chuan Ong proposed the topic, carried out the experimental study, analyzed the data

300 and writing the original draft. Hui-Juan Pan review and editing the manuscript. Noor Azhar

301 Mohamed Shazili, David Menier and Virginie Dupont supervised Meng-Chuan Ong and

302 review and editing the manuscript. Sidonie Révillon and Alexandra Connell help in

303 commenting the manuscript and interpret the results All authors read and approved the final

304 manuscript.

305

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391

392 **Figure legends**

393

394 Figure 1. Map showing surficial sediment sampling points in South Brittany waters

395 Figure 2. Concentration map for heavy metals distribution in South Brittany coastal waters

396 surficial sediment

397 Figure 3. Contamination category based on PLI values in Bay of Quiberon and Gulf of
398 Morbihan surficial sediments

399 Figure 4. Contamination category based on m-ERM-Q values in Bay of Quiberon and Gulf
400 of Morbihan surficial sediments

401

402 **Table legends**

403 Table 1. Classification of sediment samples ($\mu\text{g/g}$) based on the proposed SQGs by USEPA

404 Table 2. Classification of sediment samples based on the SQGs

Figures

Figure 1

Map showing surficial sediment sampling points in South Brittany waters

Figure 2

Concentration map for heavy metals distribution in South Brittany coastal waters surficial sediment

Figure 3

Contamination category based on PLI values in Bay of Quiberon and Gulf of Morbihan surficial sediments

Figure 4

Contamination category based on m-ERM-Q values in Bay of Quiberon and Gulf of Morbihan surficial sediments

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