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Environmental hazard assessment of a marine mine tailings deposit site and potential implications for deep-sea mining☆

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A B S T R A C T

Portmán Bay is a heavily contaminated area resulting from decades of metal mine tailings disposal, and is considered a suitable shallow-water analogue to investigate the potential ecotoxicological impact of deep-sea mining. Resuspension plumes were artificially created by removing the top layer of the mine tailings deposit by bottom trawling. Mussels were deployed at three sites: i) off the mine tailings deposit area; ii) on the mine tailings deposit beyond the influence from the resuspension plumes; iii) under the influence of the artificially generated resuspension plumes. Surface sediment samples were collected at the same sites for metal analysis and ecotoxicity assessment. Metal concentrations and a battery of biomarkers (oxidative stress, metal exposure, biotransformation and oxidative damage) were measured in different mussel tissues. The environmental hazard posed by the resuspension plumes was investigated by a quantitative weight of evidence (WOE) model that integrated all the data. The resuspension of sediments loaded with metal mine tails demonstrated that chemical contaminants were released by trawling subsequently inducing ecotoxicological impact in mussels' health. Considering as sediment quality guidelines (SQGs) those indicated in Spanish action level B for the disposal of dredged material at sea, the WOE model indicates that the hazard is slight off the mine tailings deposit, moderate on the mine tailings deposit without the influence from the resuspension plumes, and major under the influence of the resuspension plumes. Portmán Bay mine tailings deposit is a by-product of sulphide mining, and despite differences in environmental setting, it can reflect the potential ecotoxic effects to marine fauna from the impact of resuspension of plumes created by deep-sea mining of polymetallic sulphides. A similar approach as in this study could be applied in other areas affected by sediment resuspension and for testing future deep-sea mining sites in order to assess the associated environmental hazards.

1. Introduction

Portmán Bay is a heavily impacted area resulting from decades of metal mine tailings disposal that lasted until 1990. Minerals extracted in the Portmán mining district were mainly pyrite (FeS_2), galena (PbS) and sphalerite (ZnS), which were mechanically treated for concentration of metals, with about 95% of mine tailings waste generated (Martínez-Sánchez et al., 2008; Oyarzun et al., 2013). About 60 Mt of tailings were dumped into the sea, moving the shoreline seaward about 500–600 m and reaching the continental shelf off Portmán Bay (Manteca et al., 2014). The mine tailings deposit has a maximum thickness of about 14 m and is composed of fine sediments highly enriched with metals (mainly Fe, Zn, As and Pb, with metal concentrations 10 to 60 times higher than coastal sediments in the Mediterranean Sea). Above the deposit there is a thin layer of approximately 10–20 cm of coarse sediments reworked by natural (waves) and anthropogenic (bottom trawling)

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processes, with metal concentrations 10 to 20 times higher than unpolluted sediments (Cerdà-Domènech et al., 2016).

The resuspension of contaminated sediments may alter their physical and chemical characteristics, such as redox potential, pH, dissolved oxygen, potentially triggering desorption and remobilizing contaminants, affecting their mobility, bioavailability and increasing the risk of negative effects to marine fauna and ecosystem health (Bocchetti et al., 2008; Ondiviela et al., 2012). Therefore, the assessment of the impact of contaminated marine areas, such as mine tailings deposits should be investigated in different environmental matrices (sediment, water and biota) combining information from the chemistry and ecotoxicological impact, integrating data from bioavailability, bioaccumulation and biomarker responses and from ecotoxicological bioassays on bioindicator species (Viarengo et al., 2007). Biomarkers are known as important early warning signals of adverse effects, usually responding in the sub-lethal toxicity range of single or mixture of contaminants (Cajaraville et al., 2000; Annicchiarico et al., 2007; Taylor and Maher, 2016). Nevertheless, it is acknowledged that confounding factors, such as seasonality or reproductive cycle, may affect the biomarkers sensitivity, highlighting the importance of the adequate selection of bioindicator species individuals and experimental design (including controls) to allow comparability and meaningfulness of results. The integration of different quality Descriptors to assess the impact on biota and ecosystem functioning is required by the Descriptors 8 and 9 of the Marine Strategy framework directive (European Commission, 2008). The quantitative weight of evidence (WOE) model (Sediqualsoft), is considered to be a promising tool to assess the environmental hazards and ecological risks since it integrates data from the sediment chemistry, bioaccumulation, biomarkers responses and toxicity bioassays (Piva et al., 2011; Benedetti et al., 2012, 2014; Regoli et al., 2014; Bebianno et al., 2015).

The ore type exploited in the Portmán mining district, for a certain extent, is similar to that present in mid-ocean ridges and hydrothermal vent sites (ISA, 2002; Martínez-Sánchez et al., 2008; Oyarzun et al., 2013; Canals et al., 2016). Also, the hydrodynamics of the bay are low energy, somehow similar to the deep sea, being a suitable shallow-water analogue to investigate the potential impacts of deep-sea mining (Canals et al., 2016). In this sense, it is a unique place to conduct sediment resuspension experiments on a deposit of sulphide mining by-products, investigating the chemical and physical behaviour of metal loaded sediments and their ecotoxicological effects to marine organisms.

In the present study, a transplant experiment was carried out to assess the short-term effects of sediment resuspension on caged mussels (*Mytilus galloprovincialis*). Metal accumulation and biomarkers responses were analysed in mussel tissues and combined with the results from the sediment chemistry and toxicity bioassay. These were then integrated in the WOE elaboration to provide specific hazard indices for each typology of data before their overall integration to classify the hazard for the different areas and assess the impact of sediments resuspension in Portmán Bay.

2. Materials and methods

2.1. Sediments resuspension experiment and sampling sites

In the summer of 2014, the MIDAS-Portmán research cruise was conducted in Portmán Bay and in its adjacent marine area (Murcia, SE Spain) on board of the Spanish research vessels R/V Ángeles Alvariño and R/V Ramon Margalef. Transects of bottom trawling off Portmán Bay (Fig. 1) were carried out to resuspend the sediments and originate plumes, being usually less than 10 m in height, with a variable though relatively quick decline and limited dispersal, and a maximum tracking time for a given plume of about 4 h (Canals et al., 2016). Before the resuspension events, a transplant monitoring experiment was carried out with caged mussels *M. galloprovincialis* obtained from a mussel farm (Cademar) located on the Ebro Delta. Mussels (length 5.0–6.5 cm; width 1.7–3.5 cm; wet weight 20–37 g) were deployed at about 3 m above the seafloor in the following three sites (Fig. 1): off the mine tailings deposit area (Mooring_UPM2, hereafter "O", 37° 32.713′ N 0° 50.684′ W, 57 m); on the mine tailings deposit without the influence from the resuspension plumes (Mooring_UPM3, hereafter "B", 37° 34.553′ N 0° 51.563′ W, 17 m); under the influence of the artificially generated resuspension plumes (Mooring_UPM1, hereafter "P", 37° 34.177′ N 0° 51.386' W, 42 m). After 6 days of exposure, cages were retrieved on board and mussels from each site were immediately dissected and tissues (gills, digestive gland and mantle) were flash frozen and preserved at −80 °C for chemical and biomarker analyses. Surface sediment samples (top 1 cm) were also collected at the same sites and frozen at −20 °C until further analysis.

2.2. Sediments grain size analyses

Sediment grain size was determined using a Coulter LS230 Laser Diffraction Particle Size Analyser. Samples were oxidized with a 10% H_2O_2 solution to remove organic matter, and one subsample analysed and another treated with 1M HCl to remove carbonates. The total and non-biogenic grain size distribution determined and data analysed with GradiStat®.

2.3. Trace metals analyses in sediments and mussels

The concentrations of trace elements (Ag, As, Au, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Sb, Zn) in the sediment were determined after acid digestion as follows: ca. 0.5 g of dried sediment ($n = 3$) were transferred in Teflon vessels, added with 5 mL fluoridric acid and 1 mL of "aqua regia" (i.e. $HCl:HNO_3 = 3:1$) and, then, incubated at 150 °C for 90 min. At the end of the incubation, 5 mL of 10% boric acid were added and the extracts were analysed by inductively coupled plasma-atomic emission spectrometry (ICP-AES). Mercury was determined by ICP-AES exciting the element to form volatile hydride in a hydrides generation reactor, according to previously published procedure (Pohl, 2004). Standard curves were prepared in the same acid matrix used for the sediment samples. Caution was used in preparing and analysing samples to minimize contamination from air, glassware, and reagents, all of which were of Suprapur quality. Replicated measures of certified reference material (PACS-2, marine sediment reference material) and reagent blanks were used to assess precision and contamination. The analytical accuracy was routinely between 5 and 6%, and never higher than 10%. With the exception of Au, the concentrations of the same elements, were also determined in mussels tissues (gills, digestive gland and mantle) dissected from 15 individuals at each sampling site. Mussels tissues (about 0.3 g) were dried at 50 °C and digested with 5 mL nitric acid and 1 mL hydrogen peroxide in a microwave digestion system. Quality assurance and quality control were done by processing blank samples and certified reference material (CRM 278, mussel tissue). The values obtained for the certified reference materials were always within the 95% confidence interval of certified values.

2.4. Biomarkers analyses

From each site, five pools with tissues (gills, digestive gland and mantle), each of them obtained from three *M. galloprovincialis* individuals, were prepared for the analysis of the following biomarkers: oxidative stress (superoxide dismutase – SOD, catalase –