



Mobility versus retention of mercury in bare and salt marsh sediments of a recovering coastal lagoon (Ria de Aveiro, Portugal)

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ABSTRACT

During decades, mercury (Hg) was discharged into the Aveiro Lagoon. Twenty-five years after the cessation of discharges, sediment cores were collected at two areas to assess the evolution of the mercury contamination status. Vertical Hg profiles differed considerably between bare and vegetated sediments. Bare sediments contained significantly less Hg ($< 8 \text{ mg kg}^{-1}$) than historical data of 1995 (up to 40 mg kg^{-1}), probably resulting from erosion. Salt marsh sediments were marked by a Hg sub-surface enrichment, reaching 44 mg kg^{-1} in the site closer to the industrial discharge point. High Hg concentrations in 2007, 2011 and 2016 were found at similar sediment layers. These results emphasise the role of halophyte plants in the cycling and retention of Hg in sediments. The persistency of high Hg in the upper marsh layers highlights the complexity in the recovery of historically contaminated marshes and the vulnerability to modifications in hydrology associated with climate changes.

1. Introduction

In the past decades, domestic and industrial effluents have been discharged into urbanized estuaries. The resilience of trace elements in the environment and their affinity for sediments, have resulted in persistent contamination in sites with historical anthropogenic inputs (OSPAR, 2004; Hissler and Probst, 2006). Several environmental impacts have been registered, such as loss of recreational value, reduced biodiversity, fish and other food contamination, and under severe conditions, human health problems (Hylander and Goodsite, 2006). After cessation of the contamination source, commonly the closure of mines or industries, artificial remediation often has been performed, such as dredging or capping of the contaminated sediments with inert materials. Alternatively, natural remediation takes place over time, normally through the progressive deposition of particles with lower contamination, leading to the burial of contaminated sediment layers with time (Wang et al., 2004; Ridal et al., 2010). A key issue regarding natural attenuation, and one with environmental implications, is whether the contaminant effectively stabilizes within the sediments. Despite the complex biogeochemical processes in sediments, trace elements derived from human activities are usually retained in sediment

layers correspondent to the time of major inputs, providing information on the chronology and intensity of the anthropogenic sources in the area (Santschi et al., 2001; Válega et al., 2008; Outridge et al., 2011; Álvarez-Vázquez et al., 2017). Sediment properties, such as grain size, mineralogy and chemical composition may influence the contaminant binding capacity (Alloway, 1995). Binding mechanisms often involve iron and manganese oxides and sulphides (Turner, 2000). Temperature, redox potential, pH and salinity influence those processes (Ullrich et al., 2001; Celo et al., 2006). Diffusion derived from gradient concentrations and advection of solutes and particles driven by hydrodynamics, benthic organisms and halophyte plants facilitate the remobilization of trace elements and the exchanges with the water column (e.g., Berner, 1980; Boudreau, 1997). The post-mobilisation process may hence attenuate the sediment record of anthropogenic inputs. The recovery of historically contaminated areas is therefore a complex process involving numerous variables (Wang et al., 2004).

The Aveiro Lagoon also called “Ria de Aveiro” (Fig. 1) is one of the most mercury-contaminated systems in Europe (Válega et al., 2008), as a result of continuous discharges of mercury from a chlor-alkali industry between 1950 and 1994 (Pereira et al., 1998b). During this time-lapse, it was estimated that the discharge of 33 tons of mercury into the

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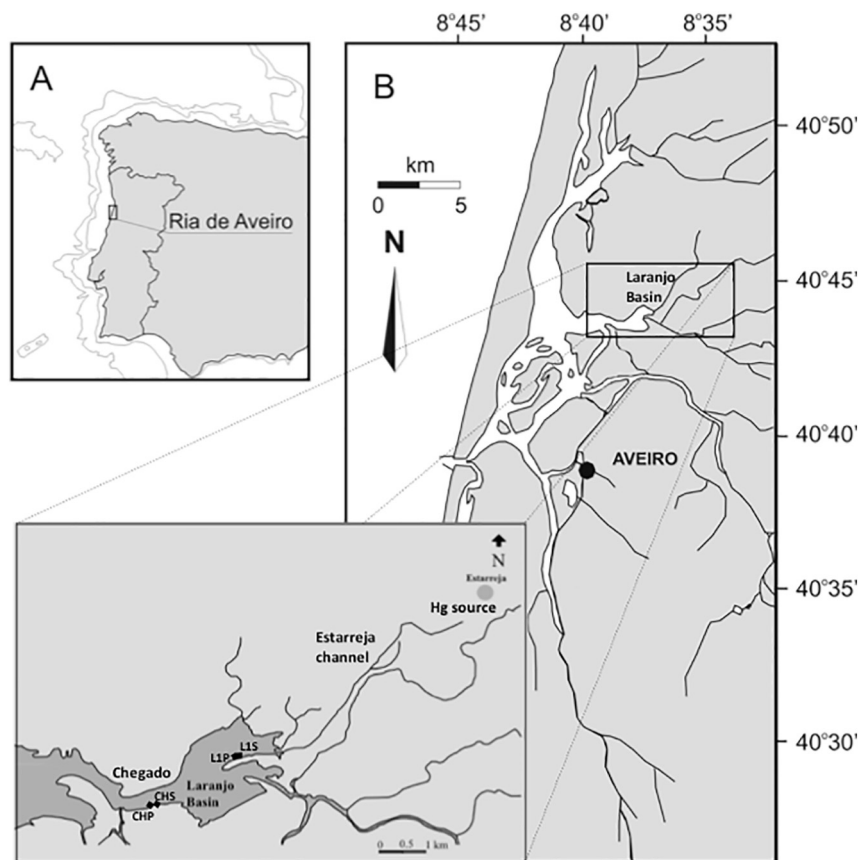


Fig. 1. Schematic plan of the Aveiro Lagoon and location of the sampling sites.

Laranjo, a 2 km² inner bay of the Aveiro Lagoon (Pereira et al., 1998b). Shallow intertidal plains and slow water currents promoted the sedimentation and burial of contaminated fine particles in the Laranjo (Pereira et al., 1998b). While a well-defined contamination gradient in relation to Hg source was found in surface sediments (Coelho et al., 2005), the highest concentrations were found in deeper sediment layers, most likely corresponding to circa 1985, when the industrial production reached the maximum (Pereira et al., 1998a; Válega et al., 2008). In a recent work Coelho et al. (2014) related the observed high suspended particulate mercury concentrations with the bottom erosion of the most contaminated sediment layers.

Various studies of mercury in wetlands and salt marshes have been performed in the last decades (e.g., Kraus et al., 1986; Weber et al., 1998; Kongchum et al., 2006). Results pointed to high Hg concentrations in the rhizosphere (De Sousa et al., 1999), low translocation into the aerial parts of plants (Weis and Weis, 2004), and low release of Hg (0) from leaves during transpiration (Windham et al., 2001). Canário et al. (2007) and Canário et al. (2010) showed high accumulation of total mercury and methyl mercury in sediments colonised by the halophyte plants *Sarcocornia fruticosa*, *Halimione portulacoides*, and *Spartina maritima* in three estuaries (Tagus, Sado and Guadiana) with high, moderate and low contamination. Moreover, belowground biomass contained higher Hg concentrations than root sediments, which implies that litter derived from belowground parts of the plants is a vehicle to the transfer of Hg to vegetated sediments.

The aim of the present work is to assess the relevance of Hg retention in salt marshes of Aveiro lagoon after cessation of the anthropogenic inputs. Depth profiles of total Hg were measured in bare sediments and sediments colonised by *H. portulacoides* in two areas of the inner Laranjo bay, where discharges occurred in the past. Mercury concentrations and depth profiles were compared with historical data, and variation over two decades was examined to test the recovery of

contaminated bare sediments and salt marsh sediments.

2. Materials and methods

2.1. Samples

Sediment cores were sampled in April 2016 in two areas 1.5 km apart: Laranjo and Chegado, (Fig. 1). The sampling areas were chosen to match previous sampling campaigns in order to allow the comparison of mercury sediment profiles with available historical data. Two sediment cores were sampled at each area, one in sub-tidal zones and another in salt marsh colonised by *H. portulacoides*. The correspondent GPS coordinates were: Site 1 – salt marsh sediment at Laranjo (L1P) - 40 43' 44" N/8 37' 01" W; Site 2 – bare sediment at Laranjo (L1S) - 40 43' 46" N/8 36' 58" W; Site 3 - salt marsh sediment at Chegado (CHP) - 40 43' 20"N/8 37' 56" W; Site 4 – bare sediment at Chegado (CHS) - 40 43' 21"N/8 37' 57" W. Sediments were collected with the help of a 100-cm corer sampler. The sediment cores were immediately withdrawn from the sampler and placed in clean polyethylene bags, to be transported to the laboratory. After arriving at the laboratory, each core was sectioned in 1 cm at the top and then every 4 cm depth. After sectioning, sediment samples were oven-dried at 50 °C until completely dry. Dried sediments were then disaggregated, homogenized and passed through a 1 mm sieve to remove roots, stones, and other macrodetritus. Finally, sediment aliquots were taken for mercury, organic matter content and grain size analysis.

2.2. Analytical determinations

2.2.1. Sediment fine fraction

The grain size analysis of the sediments was determined by wet sieving. An initial mass of sediment was weighed and then sieved

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