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Heterogeneous distribution of polycyclic aromatic hydrocarbons in surface sediments and red mullet along the Spanish Mediterranean coast



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ABSTRACT

The spatial distribution of polycyclic aromatic hydrocarbon (PAH) concentrations was characterised in surface sediments and red mullet from eleven Iberian Mediterranean coastal areas. Mean PAH concentrations ranged from 28 to 1006 and from 3 to 40 $\mu\text{g kg}^{-1}$ d.w. in sediment and red mullet muscle, respectively. The highest PAH concentrations in sediments were detected close to main ports and urban nuclei. However, concentrations of PAHs in red mullet showed little correspondence with PAH concentrations of the sediments due to its metabolic capacity. Phenanthrene was the predominant homologue in red mullet, whereas fluoranthene, pyrene and benzo(b)fluoranthene were the most abundant ones in sediments.

Significant correlations between PAHs and organic carbon or fine fraction in sediments were only found in some areas. PAH concentrations in sediments were lower than environmental criteria in the majority of cases, except for benzo(g,h,i)perylene in 25% of samples from the Barcelona coastal area and for several homologues close to the port of Sagunto.

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Polycyclic aromatic hydrocarbons (PAHs) are of special concern in the marine environment due to their widespread distribution and to the mutagenic and carcinogenic properties of their homologues, among other cytotoxic properties (White, 1986; Vethaak et al., 1996; Lyons et al., 1997; Vincent et al., 1998), with some of their metabolites displaying estrogenic activity (Van Lipzig et al., 2005; Martínez-Gómez et al., 2013). Although the main source of PAHs is the incomplete combustion of organic matter at high temperatures, other sources, such as the release of oil (oil spills, tanker ballasting/deballasting operations, etc.) or diagenetic processes (Neff, 1979), can also have a significant impact on the environment. These compounds are ubiquitous marine sediment contaminants as a consequence of their continuous access to this system through atmospheric deposition, transport in particulate and dissolved phases in groundwater and rivers, urban or industrial wastewater discharges and maritime transport spills, amongst others. In the environment, PAHs tend to be sorbed by particulate material (either biotic or abiotic) as a consequence of their hydrophobicity (high log Kow) (Pignatello and Xing, 1996), showing concentrations in particulate material that are often several orders of magnitude greater than dissolved levels (Baumard et al., 1998b). Consequently, as the bioaccumulation of a pollutant is governed by its bioavailability, organisms are often enriched in

low molecular weight PAHs in relation to sediment (Baumard et al., 1998b). Marine sediments can act as both a sink and a source of pollutants in the marine environment. If particles are ingested or the sediment-associated PAHs are otherwise released, they may be transferred to biota (Forbes and Forbes, 1998; Eggleton and Thomas, 2004).

The distribution of PAHs in fish tissues is governed by diet composition, biotransformation processes and above all their bioavailability (Baumard et al., 1998c). Thus, since vertebrates have a higher capacity to metabolize and excrete PAHs than molluscs, PAHs will tend to bioaccumulate more in the latter than in the former (Meador et al., 1995). In fact PAH levels in bivalves are commonly used as a bioindicator of their presence in the coastal water column (Baumard et al., 1998a, 1999; León et al., 2013a,b). PAH levels in benthic fish tissues contribute to our knowledge of the bioaccumulation pattern of these compounds in different fish species and may provide relevant information for the associated human consumption risks of contaminated fish (Dhananjayan and Muralidharan, 2012).

The characterisation of PAHs distribution and their temporal trends in sediments and biota are included in the international marine monitoring programs (MED POL, OSPAR, etc.). Within that context, a network to assess chemical pollution (trace metals, organochlorinated pollutants and polycyclic aromatic hydrocarbons) in sediment and marine target species (mussels and red mullet) has been developed over the last decade along the Iberian

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Mediterranean coast (Fernández et al., 2010; Martínez-Gómez et al., 2012; León et al., 2013a). The characterisation of pollutant temporal trends in sediments requires the prior identification and selection of adequate depositional and representative areas, such as bottoms undisturbed by anthropogenic activities, a high fine fraction percentage and a homogeneous content of organic matter in sediments. A pilot study (2006–2008) was conducted to characterise and identify suitable sediment sampling zones along the Spanish Mediterranean coast for future temporal trend studies, which are of particular relevance in marine areas with low sediment deposition rates, such as the one in question. In this study PAH concentrations in sediments and muscle tissue of the benthic fish red mullet (*Mullus barbatus* L., 1758) were determined in 11 areas of the Spanish Mediterranean coast from Barcelona to Málaga, including the main hotspots and the areas of influence of the major industrial and urban nuclei, ports and rivers of this Iberian coastal area. The main goals of the study were: (a) to characterise PAH distribution in surface sediments and red mullet along the Spanish Mediterranean coast, (b) to determine the PAH biota sediment accumulation factor in red mullet in relation to sediment in spring and autumn, (c) to perform an environmental assessment of the PAH concentrations detected in sediments and (d) to identify the most suitable depositional areas of the Spanish inner shelf to be used for sediment temporal trend studies and consequently for integrative assessment monitoring purposes.

A PAH mixture containing phenanthrene, anthracene, benz[a]anthracene, fluoranthene, chrysene, benzo[b]fluoranthene, pyrene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-c,d]pyrene, benzo[g,h,i]perylene and dibenzo[a,h]anthracene in acetonitrile (purity > 99%) was supplied by Dr. Ehrenstorfer (Augsburg, Germany). Benzo[e]pyrene and 2-methyl-chrysene standards in acetonitrile were also supplied by Dr. Ehrenstorfer (Augsburg, Germany). Anhydrous sulphate and aluminium oxide were supplied by Merck (Darmstadt, Germany). Methanol, water and acetonitrile gradient grade for liquid chromatography were also supplied by Merck.

Eleven sampling areas along the Iberian Mediterranean coast were considered in this study (Fig. 1 and Table 1), in order to

identify depositional areas valid for temporal trend studies. Selection of the areas was based on previously existing information of surface sediment granulometry and coastal location, including known representative polluted areas at sub regional scale. Barcelona, Tarragona, Valencia and Cartagena are identified as priority pollution hotspots in the Mediterranean Sea (UNEP/MAP/MED POL, 2005), being under the influence of significant urban, industrial and harbour activities, and of the inputs of different rivers such as the River Llobregat in Barcelona or the River Turia in Valencia. The Ebro Delta has been also identified as a priority pollution-sensitive area in the Mediterranean Sea (UNEP/MAP/MED POL, 2005), since it is an area of natural and socio-economic value under the influence of industrial and agriculture activities located in the drainage basin of the River Ebro. The second group of selected areas corresponds to areas situated close to other urban and industrial nuclei that also have ports with a relevant degree of activity but are not considered as hotspots, such as Málaga, Almería, Castellón and Sagunto. Finally, the Santa Pola Bay and Cullera areas come under the influence of the Segura and Júcar river mouths respectively, and can be influenced by the activity associated with a major urban nucleus and port situated a few kilometres to the north, namely Alicante and Valencia respectively (the predominant coastal currents move in a southward direction in these areas).

The sediment and red mullet sampling campaigns (Table 1) were performed from April to May 2006, September to October 2007 and October 2008 from O/V Francisco de Paula Navarro (IEO), and from O/V Emma Bardán (Spanish Fisheries Department) in June 2008 (Sagunto area). Only the red mullet in 2007 were obtained directly from the fishing fleets of each area.

Red mullets (*M. barbatus*) and surface sediments were simultaneously sampled from eleven muddy sedimentary areas on the inner Iberian Mediterranean shelf (Fig. 1). The only exceptions were Barcelona, Tarragona and Cartagena, where red mullets were sampled in October 2008 and sediments in 2007.

Surface sediments (top 1 cm) were sampled using a box-corer in 11 areas, including two differentiated sub-areas sampled in two successive years in Ebro Delta, Valencia and Cartagena (Table 1). Each area was subdivided into 20 × 1 nautical mile square grids



Fig. 1. Sediment and red mullet sampling areas along the Iberian Mediterranean coast obtained in different campaigns performed between April 2006 and October 2008.

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