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Source characterization and spatio–temporal evolution of the metal pollution in the sediments of the Basque estuaries (Bay of Biscay)

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ABSTRACT

According to Water Framework Directive requirements, Member States must identify and analyze effects derived from human pressures in aquatic systems. As different kind of pressures can impact water bodies at different scales, analyses of spatio-temporal evolution of water bodies becomes essential in order to understand ecosystem responses. In this investigation, an analysis of spatio-temporal evolution of sedimentary metal pollution (Cd, Cr, Cu, Hg, Ni, Pb, Zn) in 12 Basque estuaries (Bay of Biscay) is presented. Data collected in extensive sampling surveys is the basis for the GIS-based statistical approach used. The implementation of pollution abatement measures is reflected in a long-term decontamination process, mostly evident in estuaries with highest historical sediment pollution levels. Spatial evolution is determined by either naturally occurring or human driven processes. Such spatial processes are more obviously being reflected in estuaries with lower historical sediment pollution levels.

1. Introduction

Estuaries are very dynamic systems where, due to their location between marine and land environments, a wide variety of ecological and physicochemical processes take place (e.g. Ridgway and Shimmield, 2002). Estuaries are crucial ecosystems in many aspects: acting as important sites for primary production (e.g. Bricker et al., 2008); providing habitat for multiple species (e.g. Lotze, 2010); regulating material exchange processes between riverine and coastal environments (e.g. Das et al., 2010); and providing goods and services (e.g. Pinto et al., 2010).

The European Water Framework Directive (WFD, 2000/60/EC) aims to achieve "Good Ecological and Chemical Status" in all the European water bodies, by 2015 (EC, 2000; Borja, 2005; Hering et al., 2010). In this sense, many authors highlight the influence of sediments in the ecological and physico-chemical processes occurring in an estuary (Borja et al., 2004a, 2009a; Casper, 2008; Magni et al., 2008; Teixeira et al., 2008; Tueros et al., 2009). As a dynamic compartment of estuarine systems, depending on physico-chemical conditions, sediments could act either as a sink for pollutants or be turned into a new pollution source for the water column (e.g. Atkinson et al., 2007). Therefore, the identification, control and removal of pollution sources become essential in order to maintain the good ecological functioning of the water bodies (EC, 2000, 2008). This will provide mechanisms for an adequate sediment management, allowing at the same time, to protect and ensure the socioeconomic objectives of those systems (Brils, 2008; Apitz, 2012).

Such management process requires a carefully designed sediment sampling scheme, minimizing as much as possible the effect of spatial and sampling variation sources (Pattersen et al., 1999). Sampling variation could be minimized by combining a good selection of sampling sites with adequate sampling practices (Demetriades and Volden, 1997). However, considering spatial variability, the implementation of the WFD requires the handling of spatial data relevant to different spatial scales (Vogt et al., 2002). Additionally, considering the long term view of ecosystem-based management approaches, the temporal trends given by monitoring programmes would enable the adaptation of decision making processes to long-term ecological goals (MacDonald et al., 2009). Nevertheless, due to the differences in the scales at which different kind of pressures are reflected (IMPRESS, 2002), small-scale sources of spatial variance (e.g. pollutant discharge point sources) could overlap the natural large-scale spatial pattern of an estuary (Caeiro et al., 2003). Hence, under large-scale monitoring programmes, there is a risk of failing in detecting such small-scale variability sources (Kiersch et al., 2010). Therefore, periodical extensive surveys, with a substantially higher amount of locations sampled (Belzunce et al., 2001; Dean et al., 2007), will help detecting the variability derived from different spatial-scale processes. In this sense, the use of Geographical Information System (GIS) tools becomes effective, as they allow the integration of information from different sources and spatial scales (Stanbury and Starr, 2000), providing a holistic view for an adequate management of water resources.

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Hence, the approach used in this contribution, has as main objective to carry out a long-term spatio–temporal evolution analysis of the metal pollution affecting the sediments of the Basque estuaries (Bay of Biscay). For this investigation, extensive sampling surveys and GIS capabilities have been combined, considering: (i) the effect of pollution abatement measures and human pressures in the spatio–temporal variability of estuarine metal contamination; and (ii) variations occurred in the metal stocks (understood as the mass of metal fixed to sediments) during the different campaigns.

2. Materials and methods

2.1. Study area

The Basque coast is located in the southeastern Bay of Biscay and has an approximate length of 150 km (Fig. 1). Due to its geographical location, it is considered a high energy environment (González et al., 2004; Galparsoro et al., 2012). It is drained by 12 main torrential rivers, which are responsible of supplying 1.57×10^6 t yr^{-1} of suspended material into the Bay of Biscay (Uriarte et al., 2004a). Basque estuaries are strongly differentiated in terms of basin size, hydrological, morphological and dynamic features (Borja et al., 2006) (see Table A1 in Supplementary material). Although all of them can be classified as mesotidal systems, some of them show some characteristics of macrotidal estuaries (Valencia and Franco, 2004).

Historically, industrial concentration and population density have been the main drivers producing the most important pressures in these systems: water and sediment pollution; intertidal losses; and shoreline reinforcement (Borja et al., 2006). For many years, untreated domestic and industrial wastewaters have been directly dumped into the estuaries, degrading seriously the environmental quality of the area (Cearreta et al., 2004; Borja et al., 2006). Nevertheless, an overall improvement has been observed in the quality of these systems over the last years (Borja et al., 2009b, 2010; Pascual et al., 2012). Such improvements are mainly related to: (i) a decrease of the most polluting discharges, following the general decay and the changing practices in the heaviest industrial activities (Belzunce et al., 2004a,b; Leorri et al., 2008); and (ii) the diversion of wastewater discharges to coastal areas derived from the implementation of water treatment schemes (Franco et al., 2004; Tueros et al., 2009).

2.2. Sample collection and analysis

2.2.1. Sampling strategy

In the late 1990's the Basque Water Agency established an extensive monitoring programme to determine the spatial distribution of the sediments quality within the Basque estuaries. Hence, two surveys were carried out in a 11-year interval. Barbadun, Ibaizabal and Lea estuaries were sampled in 1998 and 2009; Butroe, Oka and Artibai in 1999 and 2010; Deba, Urola and Oiartzun in 2000 and 2011; and Oria, Urumea and Bidasoa in 2001 and 2012 (see Fig. 1, for locations). During each survey a total of 359 samples were collected and analyzed for sedimentological parameters whilst the metal content was determined for 128 of them. Additionally, in order to get a more complete spatial coverage, data from the *Littoral Water Quality Monitoring and Control Network* (Borja et al., 2004b) corresponding to the sampling years of each system were also included in the present analysis.

Intertidal sediments were sampled by hand, whilst sampling of subtidal samples was performed on board using either Day or Van Veen grabs. In all cases, the upper 10 cm of sediments were collected, retained in plastic bottles, transported to the laboratory and stored at 4 °C until analysis.

2.2.2. Sample analysis

Techniques used in the determination of the grain size distribution of the samples varied depending on the fine content of the sediments. Grain size of samples with low fine sediment content (<10%) was determined by dry sieving following Folk (1974); and, grain size distribution of samples with high contents of fine sediments (>10%) was determined by Laser Diffraction Particle Size Analyser (LDPSA). The mud content values for those samples measured by LDPSA, were transformed following Rodríguez and Uriarte (2009), due to the underestimation of the finest fraction of the sediment given by the LDPSA method (Campbell, 2003; Di Stefano et al., 2010).

Organic Matter (OM) content of the sediments was determined by loss of weight on ignition at 450 °C during 6 h, according to

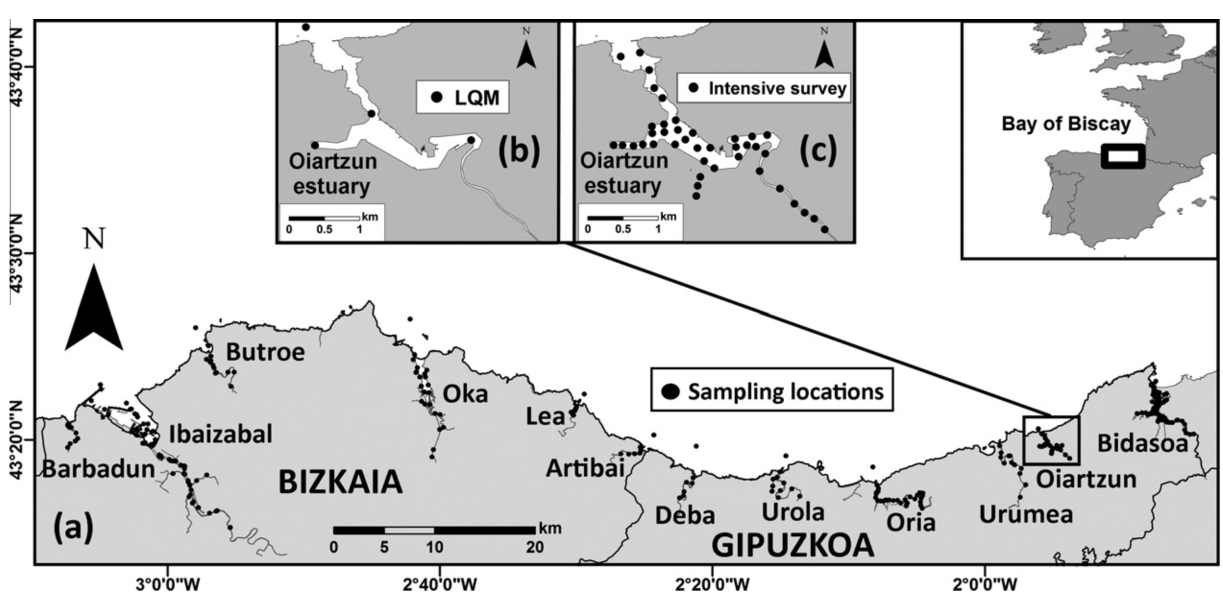

Fig. 1. (a) Study area and sampling locations in Basque estuaries, within the Bay of Biscay. Examples of different sampling approaches are also shown for the Oiartzun estuary: (b) sampling points under the *Littoral Water Quality Monitoring and Control Network* (LQM); and (c) sampling points of the intensive sediment characterization survey.