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Do pharmaceuticals bioaccumulate in marine molluscs and fish from a coastal lagoon?

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ABSTRACT

The bioaccumulation of 20 pharmaceuticals in cockle *(Cerastodema glaucum)*, noble pen shell (*Pinna nobilis*), sea snail (*Murex trunculus*), golden grey mullet (*Liza aurata*) and black goby (*Gobius niger*) was evaluated, considering their distribution throughout the Mar Menor lagoon and their variations in spring and autumn 2010. The analytical procedure was adapted for the different matrices as being sensitive and reproducible. Eighteen out of the 20 compounds analysed were found at low ng g^{-1} in these species throughout the lagoon. Hydrochlorothiazide and carbamazepine were detected in all species considered. The bioaccumulation of pharmaceuticals was heterogeneous in the lagoon, with a higher number of pharmaceuticals being detected in fish (18) than in wild molluscs (8), particularly in golden grey mullet muscle (16). B-blockers and psychiatric drugs were preferentially bioccumulated in fish and hydrochlorothiazide was also confirmed in caged clams. The higher detection frequency and concentrations found in golden grey mullet suggested that mugilids could be used as an indicator of contamination by pharmaceuticals in coastal areas. To the best of our knowledge, this is the first study that shows data about hydrochlorothiazide, levamisole and codeine in wild marine biota.

1. Introduction

In recent years, the number of studies reporting the occurrence of emerging contaminants in the marine environment has increased as a consequence of anthropogenic activities worldwide, the greater sensitivity of new analytical techniques and the rising scientific interest in this subject. The pressure and impact of organic pollutants are greater in coastal areas than in the rest of the marine environment, since these are where many human activities are concentrated and direct and indirect discharges occur (Sánchez-Bayo et al., 2011). Pharmaceuticals have frequently been detected in the aquatic environment in the range of low ng L^{-1} to low μg L^{-1} (Gros et al., 2012; Baker and Kasprzyk-Hordern, 2013; Bayen et al., 2013, 2014) and are considered as pseudo-persistent contaminants due to their degradation rate being lower than their access rate (Daughton, 2002). In the marine environment, the site conformation and hydrodynamic conditions largely determine the spatial distribution of marine pharmaceutical concentrations (Arpin-Pont et al., 2014) and they have predominantly been found in seawater (Weigel et al., 2004; Pait et al., 2006; Su et al., 2007; Xu et al., 2007; Wille et al., 2010; Yang et al., 2011; Zhao et al., 2011; Zou et al., 2011; Zhang et al., 2012, 2013; Zheng et al., 2012; Bayen et al., 2013; Rodríguez-Navas et al., 2013; Bayen et al., 2014; Xu et al., 2014; Moreno-González et al., 2015), but less so in sediment (Morales-Muñoz et al., 2005; Cueva-Mestanza et al., 2008; Landford and Thomas, 2011; Pintado-Herrera et al., 2013, 2014; Stewart, 2013; Na et al., 2013; Moreno-González et al., 2015).

Due to the general hydrophility of pharmaceuticals, their bioaccumulation potential can be considered irrelevant according to the criteria for accumulation proposed by the OECD, namely log K_{ow} (octanol-water partition coefficient) > 3 (OECD Guideline 315, 2008, Zenker et al., 2014). However, several authors reported that compounds with log K_{ow} < 2 can show high bioaccumulation (Le Bris and Pouliquen, 2004; Na et al., 2013), as has also been observed for pharmaceuticals in freshwater fish (Connors et al., 2013). In fact, the bioaccumulation of pharmaceuticals is not only determined by chemical lipophilicity but recent studies have also clearly pointed out other processes that participate in uptake and bioaccumulation, such as effects on animal homoeostasis (Van der Oost et al., 2003; Tierney et al., 2014; Daughton and Brooks, 2011; Meredith-Williams et al., 2012; Franzelliti and Fabbri, 2014), inhalational exposure as primary uptake (Du et al., 2014) and metabolization by gills (Gómez et al., 2010, 2011). Environmental pH can play an important role on their bioaccumulation since these

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