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Presence of pharmaceuticals in the Lis river (Portugal): Sources, fate and seasonal variation

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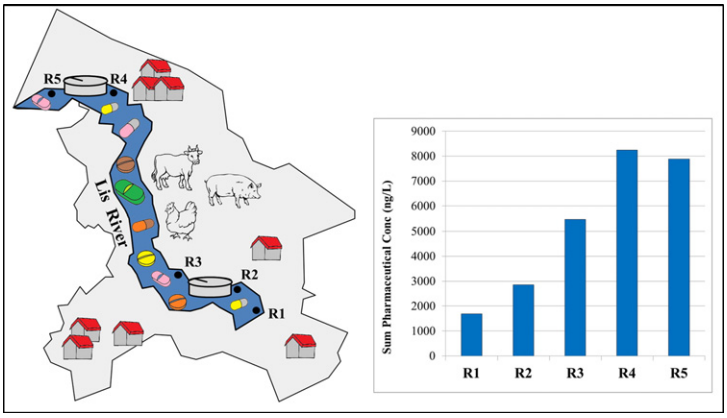
HIGHLIGHTS

- 20 pharmaceuticals and their human metabolites were detected in the Lis river.
- Ibuprofen, acetaminophen and hydroxyibuprofen showed the highest concentrations.
- 100% detection in river for carbamazepine, fluoxetine, ibuprofen, salicylic acid, ketoprofen
- WWTP effluents were identified as the most marked source of contamination.
- NSAIDs/analgesics showed the highest input from WWTPs (up to 2208 mg/d/1000 inhabitants).

GRAPHICAL ABSTRACT



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ABSTRACT

The occurrence of 33 pharmaceuticals and metabolites was evaluated along the Lis river and in the influents and effluents of two wastewater treatment plants (WWTPs) located along the river. Results indicate that pharmaceuticals, such as ibuprofen, ketoprofen, carbamazepine and fluoxetine, and the metabolite salicylic acid are widespread along the Lis river, showing 100% of detection frequency, at levels up to 1.3 μg L^{-1}. The number of molecules detected increased along the river, with 11 molecules in the source, 15 upstream WWTP 1, 16 downstream WWTP 1 and upstream WWTP 2 and 19 downstream WWTP 2. The highest concentrations were often found downstream near the river mouth. Different possible sources of contamination of the Lis river were identified, namely WWTP effluents, untreated wastewaters and livestock production. Nevertheless, the discharge of WWTP effluents appeared to be the most pronounced, given that, in general, it was noticed an increase in the concentration of pharmaceuticals downstream of the WWTPs. WWTP effluents contributed with a total mass load of pharmaceuticals into the Lis river between 470 and 2317 mg/d/1000 inhabitants. Non-steroidal anti-inflammatory drugs/analgesics were the therapeutic group with a high contribution to the total mass load of pharmaceuticals entering the Lis river, followed by psychiatric drugs and antibiotics. No seasonal variation was

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observed for the detected concentrations of pharmaceuticals. At the levels detected in the Lis river, sulfamethoxazole, clarithromycin, azithromycin and ibuprofen showed to have potential risk for aquatic organisms. These findings show that further studies embracing different environmental compartments (water, sediment and biota) are needed, in order to evaluate the partition/distribution of pharmaceuticals, their metabolites and transformation products in the environment as well as to predict their possible impact to non-target organisms and, in a last instance, to human health.

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1. Introduction

Nowadays, there are a huge number of pharmaceuticals that can be used either in human or veterinary medicine (Arpin-Pont et al., 2016). After their uptake, pharmaceuticals and their metabolites are released in wastewaters, which are treated in Wastewater Treatment Plants (WWTP). However, they are not designed to complete remove them, allowing the entrance of pharmaceuticals and their metabolites in the environment. In fact, WWTPs have been described as one of the main routes of entrance of pharmaceuticals into the aquatic environment (Verlicchi et al., 2012). Nevertheless, other possible sources of contamination by pharmaceuticals are the direct discharge of untreated wastewaters to the environment (for instance, the leakage of septic tanks) (Pal et al., 2010), landfill leachates (Paíga et al., 2013), livestock production (Lin et al., 2008), or the use of manure or WWTP sludge as fertilizer in agricultural fields (Aust et al., 2008; Thomaidi et al., 2016).

The detection of pharmaceuticals in the aquatic environment (e.g. wastewaters, surface water, groundwater, drinking water) has been globally reported, namely in the American continent (Ferrey et al., 2015), in the European continent (Alygizakis et al., 2016; Osorio et al., 2016), in the Asian continent (Sun et al., 2016; Wu et al., 2015), in the African continent (Agunbiade and Moodley, 2016; Matongo et al., 2015) and in Oceania (Birch et al., 2015; Roberts et al., 2016).

Typically, pharmaceuticals are found in river waters at low concentrations often in $ng \cdot L^{-1}$, while in WWTP effluents they are, generally, one order of magnitude higher, at levels between some hundreds of $ng \cdot L^{-1}$ to a few $\mu g \cdot L^{-1}$. Recently several studies have been published, highlighting the occurrence of pharmaceuticals in rivers, the contribution of WWTPs to their input and some possible effects due to the exposure of aquatic organisms, namely the bioaccumulation of pharmaceuticals in their tissues (Agunbiade and Moodley, 2016; de Solla et al., 2016; Roberts et al., 2016; Ruhí et al., 2016; Sun et al., 2016).

The concentration of pharmaceuticals in the aquatic environment can be influenced by several factors, such as the flow rate of the receiving surface water, which will determine their dilution factor; partitioning to the sediments and the particulate matter; biodegradation; photodegradation; other transformation reactions (e.g. abiotic mechanisms); and uptake by biota (Lindholm-Lehto et al., 2016).

The increasing detection of pharmaceuticals in the environment is leading to a problem of growing concern, given that they might pose a threat to wildlife and also to humans via, for instance, drinking water or food chain (Santos et al., 2010). In this context, the European strategy for the water policy, implemented by the Directive 2013/39/EU, determines that it is important continuing to monitor pharmaceuticals and other emerging contaminants in surface waters, in order to gather data on the most impacted surface waters, to address the risk posed by those compounds, allowing the implementation of future measures that will help to prevent and control the risks posed by those substances (European Comission, 2013).

Although the presence of pharmaceuticals in the aquatic environment is well described in many European countries, there is still a gap of knowledge in their occurrence, fate and potential environmental risk in Portugal. Just a few studies have been published, being mainly focused in rivers from the North of Portugal (Gonçalves et al., 2013; Madureira et al., 2010; Paíga et al., 2013; Pena et al., 2007; Santos et al., 2013b). Contribution of WWTP and hospital effluents to the load of pharmaceuticals into the Portuguese environment has been also described (Pereira et al., 2015; Santos et al., 2013a; Silva et al., 2014). In this context, the aim of this study was to assess the presence of 33 pharmaceuticals belonging to the therapeutic groups of non-steroidal anti-inflammatory drugs (NSAIDs)/analgesics, antibiotics and psychiatric drugs in the Lis river (Leiria, Portugal) as well as in the influent and effluent wastewaters of the WWTPs that are located along the river. Five sampling points were selected along the river, including the source of the Lis river, upstream and downstream of two WWTPs (Olhalvas WWTP and Coimbrão WWTP) that discharge their effluents into the river. It was also our purpose to define the spatial pattern of the distribution of the selected pharmaceuticals in the watercourse; to identify possible sources of contamination; to evaluate the seasonal variation of pharmaceuticals in the Lis river; and to assess the environmental risk posed by the detected pharmaceuticals to aquatic organisms.

2. Materials and methods

2.1. Sampling site, sample collection and sample pre-treatment

The Lis river is located in the centre region of Portugal and flows into the Atlantic Ocean in the Vieira de Leiria beach. It is 39.5 km long and covers a catchment area of about 495 km^2. It is recognized that the Lis river is a highly polluted river and the problems begin in the source of the river. This basin is located in a calcareous area that favours water infiltration, and given that part of the population surrounding the Lis river it is not covered by WWTPs, their untreated wastewaters are reaching the Lis river, contaminating its watercourse. The Lis river suffers a high impact of anthropogenic activities, such as agriculture, industrial activities (e.g. tannery, mineral mining) and livestock production, mainly piggeries. In the total, there are 509 piggeries and 96 facilities for cattle production located in the Lis river basin catchment. The city of Leiria is also located on the Lis riverside, having a population of approximately 63,400 inhabitants. There are also two WWTPs (Olhalvas WWTP and Coimbrão WWTP) that discharge their effluents into the river. In this context, five sampling sites were selected along the river. Grab samples were collected in the source of the Lis river (R1), upstream (R2 and R4) and downstream (R3 and R5) of two WWTPs that discharge their effluents into the river (Fig. 1). R1 is located at approximately 5 m from the river source, while R3 and R5 are located approximately 500 m downstream of the WWTP outfall. All the river samples were collected from the surface of the river and sampling was performed on the riverside. Olhalvas WWTP (WWTP 1) and Coimbrão WWTP (WWTP 2) are located at 25 and 5 km from river mouth, respectively. Olhalvas WWTP is designed for 49,315 population equivalent (serving 21,726 population equivalent in 2014), has an average flow rate of 6250 m^3/day and receives domestic wastewaters combined with rain waters. It has primary treatment operating with harrowing and desanding, and secondary treatment with conventional activated sludge. The biological treatment is performed on two aeration tanks, in aerobic conditions, which are preceded by an anoxic reactor. Olhalvas WWTP has a hydraulic retention time (HRT) of 12.5 h and a sludge retention time (SRT) of 16.4 days. Coimbrões WWTP is designed for 248,685 population equivalent (serving 110,131 population equivalent in 2014) and receives domestic wastewaters, hospital effluents, piggeries effluents and landfill leachate, combined with rain water. It has an average flow rate of

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