



A framework for the assessment of the environmental risk posed by pharmaceuticals originating from hospital effluents



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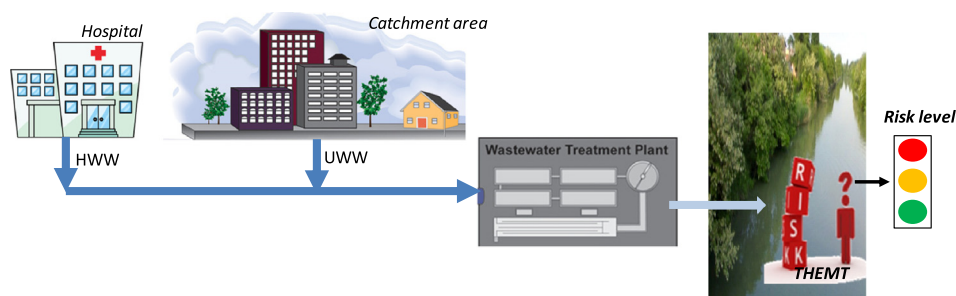
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HIGHLIGHTS

- The framework evaluates the environmental risk of PhCs originated from hospital effluent.
- The current framework includes data of 32 pharmaceuticals.
- Selected compounds derive from an analysis of those defined as priority by different studies.
- The framework considers the main characteristics of the hospital and of its catchment area.
- Ofloxacin, 17 α -ethinylestradiol, erythromycin and sulfamethoxazole are of potential concern.
- The contribution of a hospital effluent to the environmental risk is correlated to its bed density.

GRAPHICAL ABSTRACT



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ABSTRACT

The consumption of pharmaceuticals is increasing in both hospitals and households. After administration, many compounds enter the water cycle as parent compounds or their metabolites via excretion. Conventional municipal wastewater treatment plants are unable to efficiently remove all the different compounds found in sewage and, consequently, treated effluents are one of the main sources of persistent micropollutants in the environment. Hospital patients are administered relatively high quantities of drugs and therefore hospital wastewaters can consistently contribute to treatment plant influent loads, with the magnitude of environmental risk posed by pharmaceuticals originating from hospital effluents largely unknown. This study has therefore developed a framework to enable authorities responsible for hospital management and environmental health to evaluate such risk, considering site-specific information such as the contribution of human population and hospital sizes, wastewater treatment removal efficiency, and potential dilution in the receiving water body. The framework was applied to three case studies, that are representative of frequent situations in many countries, and findings demonstrated that the degree of risk posed by any compound was site-specific and depended on a combination of several factors: compound concentration and toxicity, compound removal efficiency in the wastewater treatment plant and dilution factor. Ofloxacin, 17 α -ethinylestradiol, erythromycin and sulfamethoxazole were identified as compounds of concern and might require management in order to reduce risk.

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1. Introduction

The consumption of pharmaceutical compounds (PhCs) is increasing in both hospitals and households (Van der Aa et al., 2011; Ortiz de Garcia et al., 2013; Deo and Halden, 2013). Pharmaceuticals consumed by humans are excreted in urine and feces as parent compounds or metabolites and enter the sewage system. They usually reach the wastewater treatment plant (WWTP) where they are partially removed before being discharged into surface water. Residues of pharmaceuticals in surface water have been correlated with the discharges of WWTPs (Al Aukidy et al., 2012). As the human body may only metabolize a fraction of the administered PhC, it enters the water cycle as the parent compound and/or its metabolites via excretion, mainly in urine (55–80%) and to a lesser extent the feces (4–30%) (Jjemba, 2006). The excretion rate is strictly correlated to a specific compound and to individual human characteristics, including age, gender, health status and concurrent assumption of other substances (Johnson and Williams, 2004).

Conventional municipal WWTPs are unable to efficiently remove all the different compounds found in sewage, and treated effluents are therefore one of the main sources of PhC release into the environment (Verlicchi et al., 2012b). Over the last ten to fifteen years, increasing attention has been paid to the fate and effects of PhCs in the environment and in particular in surface water bodies. PhC concentrations in the different water environments (raw and treated urban wastewater (WW) and surface water) have been extensively monitored in order to assess the removal ability of conventional treatment plants and their occurrence in the final water bodies. Some compounds were found to cause negative effects to the environment: among them diclofenac (Oaks et al., 2004) and hormones (Routledge et al., 1998).

Nevertheless, PhCs are still unregulated (emerging) compounds, and there is an ongoing debate within the scientific community regarding which PhCs to include among the substances demanding priority attention (Bottoni et al., 2010). Indeed, according to the European Community Directive, 2013/39 (2013), the anti-inflammatory diclofenac and the hormones 17 β -estradiol and 17 α -ethinylestradiol have been included in the European Watch List, while according to the US EPA, erythromycin, nitroglycerin, and 9 hormones need to be considered a priority (Richardson and Ternes, 2011).

Hospital wastewater (HWW) represents a particular concern. In the last few years it has become the object of several investigations (among them: Verlicchi et al., 2010b; Ort et al., 2010; Escher et al., 2011; Santos et al., 2013), but up to now studies dealing with it are still fewer than those referring to urban wastewater (UWW). These investigations are made difficult not only by high analysis costs, but also by the difficulties in organizing water-sampling campaigns inside health facilities. Nonetheless, according to some recent literature (Ort et al., 2010; Riazul Haq et al., 2012; Verlicchi et al., 2010a,b, 2013a), HWW represents the main source for some pharmaceuticals in terms of the PhC load generated, prompting the scientific community to question the acceptability of the general practice of discharging it into public sewers, where it is conveyed to municipal WWTPs and co-treated with UWWs (Kümmerer and Halmers, 2000; Pauwels and Verstraete, 2006; Verlicchi et al., 2010a, 2010b). Initially, the discussion centered on the concentrations of regulated (e.g. organic substances, N and P compounds, and microorganisms) and unregulated (residual of PhCs) pollutants in both hospital and urban WWs (Kümmerer and Halmers, 2000; Pauwels and Verstraete, 2006). The focus then shifted to the evaluation of consumption and load of selected PhCs (on the basis of high consumption and negative environmental effects documented by literature) produced by a hospital and its catchment area (Escher et al., 2011; Heberer and Feldmann, 2005; Ort et al., 2010; Riazul Haq et al., 2012; Schuster et al., 2008; Verlicchi et al., 2012a). In this context, Escher et al. (2011) reported that in a Swiss hospital of 338 beds 1154 kg of PhCs were consumed in 2007 and in the corresponding catchment area 18,700 kg were consumed by the resident population (250,000 inhabitants) in the same year. The cited studies estimated the relative contributions

made by the hospital to its catchment area for each compound under investigation, revealing that in some cases the hospital is indeed the main source of certain PhCs in WW, for example the antibiotics ciprofloxacin, spiramycin, clarithromycin, azithromycin and ofloxacin (Le Corre et al., 2012; Riazul Haq et al., 2012; Santos et al., 2013; Verlicchi et al., 2012a) and the lipid regulator atorvastatin (Ort et al., 2010; Verlicchi et al., 2012a).

At the same time, several research groups set out to quantify the environmental risk generated by selected PhCs in raw hospital and urban WWs, as well as in municipal WWTP effluents (Escher et al., 2011; Verlicchi et al., 2012a). Through evaluation of a compound's risk quotient (RQ), i.e. the ratio between its measured or predicted concentration and its predicted no-effect concentration (PNEC), these studies have shown that for some compounds the risk is high ($RQ > 1$) in raw WWs and remains high in the WWTP effluent. However, once the effluent is discharged into the receiving water body, its dilution with surface water may mitigate the effect of residual PhCs and the associated risk quotient may decrease (Gros et al., 2010), sometimes even to medium or low levels. Traces of PhCs were also detected in groundwater and drinking water, being this the result of a contamination of the aquifer and of water works not able to remove micropollutants such as PhCs.

All cited studies were conducted with the aid of local PhC consumption data and/or field monitoring campaigns. These types of investigations are costly and time-consuming. Therefore, in the case of construction of a new hospital, for example, a simple screening procedure, able to provide a rough estimation of the potential impact of the PhCs in its effluent on the local environment, would be valuable for the authorities and decision-makers responsible for hospital management and environmental protection. To this end, the aim of this study is to provide the authorities responsible for hospital management and environmental health with a simple approach (a framework) to evaluate the potential environmental impact of the hospital effluent, taking in consideration site-specific information such as the number of hospital beds, the quantity of wastewater discharged in the catchment area, the removal efficiency of the local WWTP, and the available dilution of the discharged effluent in surface water body. This study also aims to assess the relative importance of PhC main sources (HWWs, UWWs) for a single compound. The case of the priority candidate diclofenac, considered a target compound for individual WWTPs, will be presented. Such information will then be discussed to demonstrate its potential to assist with options for reducing PhC risk in discharges and to highlight the need to adopt management options.

2. Materials and methods

2.1. The proposed framework

This framework is designed to estimate, for a given hospital, the potential environmental impact posed by selected PhCs in the hospital effluent in terms of their RQ ranges. It takes into consideration the characteristics of: (i) the hospital in question (number of beds, wards, water consumption), (ii) its catchment area (number of inhabitants, water demand), (iii) the treatment process used for the hospital effluent and the corresponding removal efficiencies for the selected compounds, and (iv) the characteristics of the receiving water body (dilution and hydrodynamic features).

The PhCs discussed in this study focused on the minimum number of compounds that should be considered in any study on PhC in water management. These compounds have been defined as high priority or priority substances by different research groups worldwide (Besse and Garric, 2008; De Voogt et al., 2009; European Community Directive, 2013/39, 2013; Ginebreda et al., 2012; NRMCM, 2008; Perazzolo et al., 2010; Richardson and Ternes, 2011; Roos et al., 2012; Ruel et al., 2012; Santos et al., 2013; Sui et al., 2012; Verlicchi et al., 2012b). An in-depth analysis is reported in Table SI-1 in the Supplementary information section. A total of thirty two PhCs were selected, among them six analgesics and anti-inflammatories, twelve antibiotics, one

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