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Hospital effluent: Investigation of the concentrations and distribution of pharmaceuticals and environmental risk assessment

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ABSTRACT

A study was conducted in an area in north, Italy, on the effluent of two different sized hospitals and the influent and effluent of the receiving municipal treatment plant of one of the examined hospitals. The aim was to investigate 73 selected pharmaceuticals, belonging to twelve different classes, comparing their occurrence in the effluent directly exiting the hospital with that, mixed with the local urban effluent, at the point of its entry and exit from the treatment plant.

Consistent differences were found in the concentrations of some antibiotics, analgesics and lipid regulators in the two wastewaters, confirming that hospital effluents should not be considered as possessing the same pollutant nature as urban wastewater. Furthermore, analysis of percentage contributions of the hospital to the treatment plant influent evidences that hospitals represent one of the main sources of pollutants, in particular antibiotics, receptor antagonists and lipid regulators.

Hence, an environmental risk assessment, performed on the effluent from the hospital and the influent and effluent from the treatment plant, revealed a high risk for 9 pharmaceuticals in hospital effluent and for 4 of the 9 substances in the treatment plant influent and effluent, with antibiotics being the most critical compounds in terms of contribution and potential environmental risk for the hospital.

1. Introduction

During recent years, the issue of pharmaceutical compounds (PhCs) in wastewater has become a major concern in terms of both human health and the environment. This has prompted the launch of several monitoring studies into the most commonly administered compounds in urban wastewater (Lishman et al., 2006; Santos et al., 2007; Terzic et al., 2009) and surface water (Kolpin et al., 2002).

However, a considerably smaller number of studies have been devoted to characterizing PhCs sources, mainly hospital effluents (Boillot et al., 2008; Kosma et al., 2010; Kummerer, 2001; Sim et al., 2011). In fact, in quite all countries worldwide, no distinction is usually made between these wastewaters and urban effluent, and they, along with their potentially hazardous loads, are generally discharged directly into the public sewage network and conveyed for co-treatment at the nearest municipal wastewater treatment plant (WWTP).

Nonetheless, considering the multiple research and laboratory activities carried out in these structures, as well as the treatments performed and pharmaceuticals administered and excreted within them, a wide range of concentrations of hazardous substances may be present in hospital effluent (Verlicchi et al., 2010). Hospital wastewaters are composed of the effluents of different services: kitchen, internal laundry, heating and cooling systems, laboratories, radiology departments, outpatients departments, transfusion centres and wards. Due to the nature and quantity of the micro-pollutants they harbor, such as active substances of medicines and their metabolites, chemicals, heavy metals, disinfectants, sterilizers, and radioactive markers, which are typically present at concentrations of μg/L, they should be earmarked for special consideration. Previous studies investigated the occurrence in hospital effluents of detergents, disinfectants, organic compounds (alcohols, acetone, formaldehyde, acetaldehyde, phenols) and several metals (Emmanuel et al., 2005; Boillot et al., 2008) and the proliferation of drug-resistant microorganisms (Hawkshead, 2008). The issue of PhC occurrence in hospital effluents has already been investigated by different Authors, among them Thomas et al., 2007; Gomez et al., 2006; Mahnik et al., 2007; Suarez et al., 2009; Kummerer, 2001.

It would therefore be of interest to discover the percentage contributions of PhCs from hospitals to those in the total municipal WWTP influent, in order to discover whether specific treatments for hospital effluent are necessary to reduce environmental contamination by persistent and hazardous micropollutants. To date, however, very

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little data on this topic has been reported in the literature (Beier et al., 2011; Heberer and Feldmann, 2005; Langford and Thomas, 2009; Ort et al., 2010; Thomas et al., 2007), and those studies have been conducted to a limited number of compounds.

In order to investigate the differences between hospital and urban wastewaters, an assessment of the (acute and chronic) risk posed to aquatic organisms by the two effluents would be advisable. In fact, although the ecotoxicological effect of PhCs in treated urban wastewaters has been investigated (Ferrari et al., 2003; Kostich and Lazorchak, 2008), once again, very little data is available regarding hospital effluent, and what is available generally relies on predicted, rather than measured, concentrations (Escher et al., 2011).

Therefore, in this study we set out to investigate the occurrence of 73 common PhCs from 12 different therapeutic classes in the effluent of two hospitals (medium-sized and large) in a town in the Po Valley, north Italy, and in the influent and effluent of the local municipal WWTP, which also receives and co-treats the wastewater from the larger hospital. The aims of the study were: (i) to compare the PhC concentrations discharged by the two hospitals over the same period, (ii) to evaluate the PhCs discharged by the large hospital over two different periods, (iii) to compare these concentrations with those found in the influent to the WWTP during the same period, (iv) to evaluate the contribution, in terms of the compounds detected, of the large hospital to the total influent to the WWTP, and finally (v) to assess and compare the potential environmental risk of hospital effluent and WWTP influent by evaluating the ratio between the measured environmental concentration (MEC) and the predicted no-effect concentration (PNEC) for these wastewaters.

In this way, our study attempts to provide an initial assessment of these issues with a view to comparing the chemical and ecotoxicological characteristics of hospital effluent with those of the influent to the WWTP charged with co-treating hospital wastewater.

2. Experimental materials and methods

2.1. The two hospitals and WWTP under investigation

Hospital A: it is a medium-sized hospital with 300 beds, 650 members of staff and twelve main wards. It is situated in a small urban settlement (5000 inhabitants), few km from the sea, in a coastal area that is densely populated in summertime due to tourist influx (in the peak months of July and August, the population is seven times higher than the resident one). Hospital flow rate is regularly monitored by the internal Water and Wastewater Network Managing Body. The resulting average flow rate is equal to 160 $m^3\ d^{-1}$, corresponding to a specific water consumption of about 550 $L\ bed^{-1}\ d^{-1}$.

Hospital B: it is a large hospital with 900 beds, 2000 members of staff and a total of over 50 wards and departments. It is located in the centre of a town (135 000 inhabitants) and its effluent is directly discharged into the combined sewage network, conveyed to the large municipal WWTP and co-treated with the urban WWs. Hospital B flow rate is regularly monitored by the internal Water and Wastewater Network Managing Body. The resulting average flow rate is equal to 603 $m^3\ d^{-1}$, corresponding to a specific water consumption of about 670 $L\ bed^{-1}\ d^{-1}$, and its bed density, that is the number of beds per 1000 inhabitants, is roughly 6.5.

The large municipal WWTP: designed for 120 000 population equivalent (pe), it performs preliminary treatments (screening and grit removal), a biological treatment and a final NaClO disinfection step. The biological treatment consists of a conventional activated sludge system including denitrification ($V = 4000\ m^3$) and nitrification ($V = 6100\ m^3$) steps, followed by secondary sedimentation ($V = 6000\ m^3$). It operates at a low-to-medium load, at an average hydraulic retention time of 6 h, a sludge age of 8 d and a mixed liquor concentration of approximately 3.5 $kg\ m^{-3}$. The WWTP influent flow rate is on average 28 000 $m^3\ d^{-1}$, and Hospital B contributes roughly 2% of the influent hydraulic load.

2.2. Target compounds

The 73 PhCs under investigation are reported in Table 1, grouped according to their therapeutic class. These compounds were selected due to their high prescription rates or volumes, the availability of a reliable analysis methods (Gros et al., 2006), as well as due to their occurrence and ubiquity in the aquatic environment (Bell et al., 2011; Daughton and Ternes, 1999; Fatta-Kassinos et al., 2011; Pal et al., 2010). The selected compounds represent the most consumed within their corresponding therapeutical class. It is quite evident that analgesics and anti-inflammatories are the groups most investigated, followed by beta-blockers and lipid regulators.

2.3. Sampling sites and sample preparation

Four sampling points were monitored: the effluents from Hospitals A and B and the influent and the effluent of the large municipal WWTP. Two experimental campaigns were carried out in August 2009 (summer) and in March 2010 (winter). In the first period, water samples were taken from the raw effluent of Hospital A ($n = 4$) and Hospital B ($n = 4$), while in the second one, from the effluent of Hospital B ($n = 4$) and the influent and the effluent of the large municipal WWTP ($n = 4$).

Manholes located on the property line of each hospital were selected as sampling points, based on their suitability for covering all of the sewage discharges from the facility. Portable auto samplers (Sigma 900) were used to collect samples from each sampling point.

24-hour composite water samples were collected over four days on each sampling point at a rate of one sample per hour (a total of 24 sub-samples, 125 mL each were collected over 24 h). To insure representative sampling and consistency in the estimation of the mass loadings at the differing locations, identical sampling strategies (the same sampling frequencies) were used for both Hospital B effluent and WWTP influent. Water samples were collected only in dry days in order to avoid dilution effects. Wastewater samples were collected in amber glass bottles, pre-rinsed with ultra-pure water, as 24-h composite samples. The samples were immediately transported to the near laboratory under cooled conditions (4 °C). Upon reception, samples were filtered through 0.45 μm Nylon filters (Whatman, Maidstone, UK) to eliminate suspended solid matter and then frozen until analysis (less than a week) at −20 °C. It is important to observe that the fraction of the selected pharmaceutical sorbed onto the suspended solids is removed during preparation phase and, as a consequence, the values of (measured) concentrations found correspond to the dissolved fraction of the investigated compounds.

2.4. Standards

All standard solutions used were of a high purity grade (>90%). Isotopically labelled compounds, used as internal standards, were: ^{13}C-phenacetin, fluoxetine-d_5 and flumequine from Sigma-Aldrich (Steinham, Germany), sulfathiazole-d_4 from Toronto Research Chemicals, diazepam-d_5 and phenobarbital-d_5 from Cerilliant (Texas, USA), atenolol-d_7, carbamazepine-d_{10}, ibuprofen-d_3 from CDN isotopes (Quebec, Canada) and mecoprop-d_3 from Dr. Ehrenstorfer (Augsburg, Germany).

Both individual stock standard and isotopically labelled internal standard solutions were prepared on a weight basis in methanol, except fluoroquinolones, which were dissolved in a water:methanol mixture (1:1) containing 0.2% v/v hydrochloric acid (Golet et al., 2002). After preparation, standards were stored at −20 °C.

Due to their limited stability, fresh stock solutions of antibiotics were prepared monthly, while stock solutions for the other substances were renewed every three months.