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Fate of antibiotics from hospital and domestic sources in a sewage network

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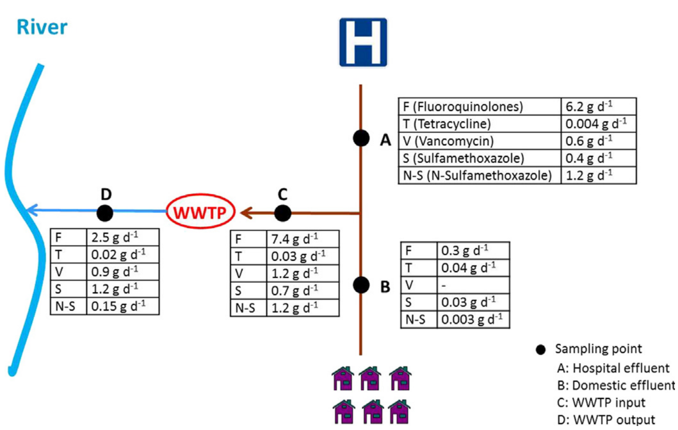
HIGHLIGHTS

- Fluoroquinolones prevail in domestic as well as hospital effluents.
- Amoxicillin, the most consumed antibiotic, is not persistent.
- WWTP treatment remains inefficient for some antibiotics such as sulfamethoxazole.
- The parent molecule is restored from its metabolite *N*-acetyl sulfamethoxazole.
- Hospital responsibility for wastewater antibiotic levels is clearly demonstrated.

GRAPHICAL ABSTRACT



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ABSTRACT

Investigation of domestic and hospital effluents in a sewage system of an elementary watershed showed that antibiotics belonging to eight classes were present with concentrations ranging from <LOQ to 50 μg L^{-1}. The compounds most often detected in the effluents were the fluoroquinolones (79–100%), the sulfonamides (86–100%) and the macrolides (79–86%). Vancomycin, strictly reserved for hospital use in France, was detected exclusively in the hospital effluent, supporting its pertinent use as a marker of hospital discharge. Beta-lactams, which are among the most frequently consumed compounds, were rarely detected in the effluents, due to their rapid hydrolysis. Out of 23 antibiotics used in veterinary and human medicine, fourteen were quantified in both the wastewater treatment plant (WWTP) input and output: erythromycin, amoxicillin, tetracycline, trimethoprim, ormethoprim, sulfamethoxazole, vancomycin and seven quinolones (flumequine, enrofloxacin, enoxacin, ofloxacin, lomefloxacin, norfloxacin and ciprofloxacin). Antibiotic concentrations in the hospital effluent (from 0.04 to 17.9 μg L^{-1}) were ten times higher than those measured in the domestic effluent (from 0.03 to 1.75 μg L^{-1}), contributing to 90% of the antibiotic inputs to the WWTP. Some molecules such as sulfamethoxazole, erythromycin and trimethoprim displayed higher concentrations after wastewater treatment due to deconjugation of their metabolites, which restores the parent molecules. For other compounds, the antibiotic elimination showed discrepancies depending on their physicochemical properties. For fluoroquinolones, the apparent removal

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processes were mainly based on adsorption mechanisms, followed by settling, leading to sludge contamination (from 13 to 18,800 μg kg^{-1} dry weight).

1. Introduction

Antibiotics are widely used in human and veterinary medicine to reduce the burden of common infectious diseases and have become essential for many medical purposes. Their total worldwide consumption was estimated to range from 100,000 to 200,000 t per year (Wise, 2002). Over the last decade, a 36% global increase in antibiotic consumption from 54.1×10^9 to 73.6×10^9 standard units was reported (Van Boeckel et al., 2014). In 2012, for the 26 European Union countries, the amounts of active substances purchased were 3400 t for humans and 7982 t for breeding animals (ECDC, 2015). In France in 2005, a high consumption of 760 t for human medicine and of 1320 t for veterinary medicine was reported (Moulin et al., 2008). This antibiotic consumption is among the highest in European countries (ANSM, 2014).

In human medicine, beta-lactams such as penicillins and cephalosporins are the main class of agents used for treating bacterial infections, accounting for 50–70% of total antibiotic use (ECDC, 2010). Next, by decreasing order of use, come sulfonamides, macrolides and fluoroquinolones, accounting for 15% altogether. Some antibiotics such as cefotaxime and vancomycin are restricted to hospital use (Vidal, 2016). In the veterinary sector, tetracyclines account for 40% of total consumption, followed by the association of sulfonamides with diaminopyrimidines (ANSES, 2016).

After administration, depending on the compound chemical properties, from 5 to 90% of the antibiotic dose may be excreted either as metabolites or as parent compounds (Kümmerer, 2009). Therefore, large amounts of antibiotics may directly enter the aquatic systems via wastewater treatment plant (WWTP) effluent discharges or via aquaculture activities (Sarmah et al., 2006). Also, the application of sludge and animal manure to agricultural fields as fertilizers may contaminate agricultural soils and lead to indirect inputs of antibiotics into hydrosystems (Wise, 2002).

Antibiotics are ubiquitous in many environmental compartments such as wastewater (Rossmann et al., 2014), surface water (Deng et al., 2016; He et al., 2015), sediment (Chen and Zhou, 2014; Matongo et al., 2015) and soil (Pan and Chu, 2016; Solliec et al., 2016). Moreover, the persistence of some antibiotics, from several days to several months, in water (Ingerslev et al., 2001), sediment or soil (Díaz-Cruz et al., 2003) has been reported. Problems have emerged during the last decade concerning substances without regulatory status such as antibiotic residues (Carraro et al., 2016; Rodriguez-Mozaz et al., 2015). Among the multiple reservoirs and pathways of antibiotic resistance dissemination, hospital, municipal sewage collectors and WWTPs are known to play an important role (Varela et al., 2016). Classical treatment in municipal WWTPs remains of limited removal efficiency for a wide spectrum of antibiotics (Verlicchi et al., 2015). For norfloxacin and ciprofloxacin, >70% of the total amount passing through the plant was ultimately found in the digested sludge (Lindberg et al., 2006). Thus, a number of antibiotic input sources to the environment that might promote the selection of antibiotic-resistant genes and bacterial strains are highlighted. Indeed, antibiotic-resistant pathogens have emerged and were disseminated among human and animal populations worldwide. Pathogens such as methicillin-resistant *Staphylococcus aureus* and beta-lactam-resistant *Enterobacteriaceae* have become a global concern (Rizzo et al., 2013). Vancomycin-resistant *Enterococci*, a leading cause of nosocomial infections, were detected in wastewater (Rosenberg Goldstein et al., 2014).

In that context, this study investigated, in both the dissolved and particulate phases of wastewaters from an elementary river basin in rural area, the 23 most commonly used antibiotics belonging to eight classes and two metabolites frequently found in effluents. The first objective was to determine the respective proportions of antibiotic contributions related to hospital medicine and family medicine, considering two types of effluents: from hospital and domestic sources. The second objective was, considering WWTP inputs and outputs, to improve knowledge on the fate of antibiotics during wastewater treatment and to provide a mass balance estimation.

2. Material and methods

2.1. Description of the study area

The transfer and the fate of 23 antibiotics were studied in the sewage network of a rural municipality of the Essonne district (91, France): Fontenay-les-Briis (inhab: 1882, population density: 194 inhab km^{-2}) located in an elementary river basin (Fig. 1). The site studied was equipped with a separate sanitary sewer system. The Fontenay-les-Briis WWTP treated wastewaters originating in both domestic (60%) and hospital (40%) sources. The 360-bed hospital was a polyvalent medical center whose discharge was controlled by flow-rate measurement. The WWTP, equipped with a combined tank (decantation and activated sludge), annually treated 155,000 m^3 of wastewater using biological processes and produced approximately 32 t of dry sludge. The mean daily flow was 425 $m^3\,d^{-1}$ of which 170 $m^3\,d^{-1}$ came from the hospital effluent and 255 $m^3\,d^{-1}$ from domestic effluent. The treatment capacity corresponded to 5000 inhabitant equivalents, while a population of 1700 inhabitants was connected, indicating that the system was clearly oversized. The flow of wastewater entering the WWTP ranged from 270 to 532 $m^3\,d^{-1}$ in 2010–2011 and the average transit time was 17 h. The annual mean decrease between input and output for biological oxygen demand (BOD5), chemical oxygen demand (COD) and suspended solids were 98%, 91% and 95.2%, respectively (corresponding to 1.6, 14.3 and 1.9 kg d^{-1} in the output, respectively), during the study period, highlighting the WWTP efficiency for these parameters. Sludge was formed by a settling process of suspended particles and the removal efficiency for the suspended matter ranged from 82% to 96%. Then sludge was stored under anaerobic conditions in an 83-m^3 silo and dried on sand beds (6×100 m^2).

Wastewaters were collected i) in the hospital sewer, ii) in the domestic sewer, iii) in the main sewer after mixing of both wastewaters flowing to the WWTP and iv) in the WWTP output (Fig. 1). They consisted to grab samples collected monthly from December 2009 to February 2011 (n = 14). Antibiotic concentrations were determined both in the dissolved and the particulate wastewater phases. Characteristics of bulk wastewater samples are indicated in the Supplementary material (Table SM.1).

2.2. Chemicals

The antibiotics studied were: sulfonamides (sulfamethazine (SMZ) and sulfamethoxazole (SMX)); quinolones (oxolinic acid (OXO), nalidixic acid (NAL) and pipemidic acid (PIP)); fluoroquinolones (flumequine (FLU), enrofloxacin (ENR), ciprofloxacin (CIP), ofloxacin (OFL), norfloxacin (NOR), enoxacin (ENO), lomefloxacin (LOM) and sarafloxacin (SAR)); tetracyclines (tetracycline (TET) and chlortetracycline (CTE)); macrolides (tylosin (TLS) and erythromycin (ERY)); beta-lactams (amoxicillin (AMO) and cefotaxime (CEF)); diaminopyrimidines (trimethoprim (TRI) and ormethoprim (ORM)); nitro-imidazoles (ornidazole (ORN)) and glycopeptides (vancomycin

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