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Antibiotics in the aquatic environments: A review of the European scenario

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ABSTRACT

The discovery of antibiotics is considered one of the most significant scientific achievements of the 20th century, revolutionizing both human and veterinary medicine. However, antibiotics have been recently recognized as an emerging class of environmental contaminants since they have been massively administrated in humans and animals and persist in the environment through a complex vicious cycle of transformation and bioaccumulation. The diffusion of antibiotics in the environment, particularly in natural water systems, contributes to the development and global dissemination of antibiotic resistance. This phenomenon is one of the most important challenges to the health care sector in the 21st century. As a result, studies on the occurrence, fate, and effects of antibiotics in European aqueous environments have increased in the last years. Nevertheless, their potential aquatic ecotoxicity and human toxicity via environmental exposure routes remain unknown. Consequently, antibiotics are not regulated through the current European environmental water quality standards, which requires evidence concerning their widespread environmental contamination and intrinsic hazard. In this context, this literature review summarizes the state of knowledge on the occurrence of antibiotics in the different aqueous environmental systems across the Europe, as reported since 2000. Relating this subject to antibiotic consumption and their dynamic behavior in the environment, the acquired insights provide an improved understanding on aquatic pollution by antibiotics to outline the European scenario. Moreover, it addresses challenges, prospects for future research, and typical topics to stimulate discussion.

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1. Introduction

In recent years, the availability of robust and sensible instrumental methods of chemical analysis have enabled studies on the occurrence of many emergent contaminants (ECs) in environmental systems at low concentrations (Zwiener and Frimmel, 2004; Farré et al., 2012; Fischer et al., 2012). ECs comprise an extensive variety of chemical substances characterized by wide distribution and persistence in the environment due to their massive used in the everyday for different purposes. Pharmaceuticals and personal care products (PPCPs), plasticizers, flame retardants, and pesticides are the main ECs categories (Sauvé and Desrosiers, 2014). The fate and the potential effects and risks of these compounds on ecosystems and human health at such concentrations have been a topic of increasing concern and research worldwide. This interest justifies the reasonably large number of reviews on the analytical determination (e.g. Gros et al. (2006a), Hogenboom et al. (2009), Albero et al. (2015), and Subedi et al. (2015)), occurrence, distribution, and risks of ECs in environment, particularly in aquatic compartments (e.g. Sørensen et al. (1998), Heberer (2002), Ritter et al. (2002), Hernando et al. (2006), Kümmerer (2009a), Lapworth et al. (2012), Bu et al. (2013), Li (2014), Meffe and de Bustamante (2014), and Pal et al. (2014)). However, little is currently known about the potential toxicological and ecological impact of ECs in aquatic ecosystems (Stuart et al., 2012; Bellenger and Cabana, 2014; Petrie et al., 2014), which explains the fact that most of these compounds are uncovered by current existing regulations on water quality (EC, 2000, 2001, 2008, 2013; US EPA, 2014; EC, 2015a). Therefore, further scientific research on their effects on environment and human health are required in order to the competent authorities establish effective and appropriate regulation and monitoring strategies for the substances identified as priority or particularly relevant in the field of water (Lepom et al., 2009; Murphy et al., 2012).

Among pharmaceuticals, antibiotics have received particular attention since the late 90s due to correlations between the development and rapid expansion of antibiotic resistance and their total consumption and occurrence in the environment (Levy, 1997; Barbosa and Levy, 2000; Aryal, 2001; Levy, 2002). Municipal, agricultural, and industrial wastewater are the major entrance sources and pathways of antibiotics and their by-products (BPs), namely metabolites and transformation products (TPs), in the environment, since substantial amounts (30% to up 90%) of antibiotics administered to humans and animals are excreted into waste stream via urine and feces, largely unmetabolized, and conventional wastewater and recycled water treatments are only partially effective in their removal or degradation (Xu et al., 2007; Teixeira et al., 2008; Leung et al., 2012; Kim et al., 2013; Santos et al., 2013; Rossmann et al., 2014; He et al., 2015; Mozaz et al., 2015; Xu et al., 2015). As a result, depending on mobility and the persistence in the soil-water environment, antibiotics and their BPs may reach surface waters and groundwaters, and, potentially, drinking waters. The detected concentrations generally range from ng L^{-1} to μg L^{-1} according to the aqueous environment matrices (Barnes et al., 2008; Focazio et al., 2008; Tamtam et al., 2008; Jiang et al., 2011; Homem et al., 2014; Tong et al., 2014). The determination of the environmental contamination levels by antibiotics and their BPs is fundamental to improve the current state of knowledge on their source pathways, transport, fate, and effects in the environment (Ternes et al., 2015).

Review articles on sources, occurrence, fate, and risks of antibiotics in aquatic environments have been published since the late 20th century. These papers commonly provided a global perspective on the subject, summarizing the information of studies conducted worldwide and highlighting the lack of comprehension on the potential toxicological consequences of antibiotics in ecosystems (e.g. Hirsch et al. (1999), Kümmerer (2003), Sarmah et al. (2006), Baquero et al. (2008), Kemper (2008), Martinez (2009), Kümmerer (2009b), Kümmerer (2009c), Milić et al. (2013), and Manzetti and Ghisi (2014)). Moreover, some publications introduced a critical review on the removal efficiency of antibiotics from wastewater through different treatment processes (e.g. Le-Minh et al. (2010), Homem and Santos (2011), Oller et al. (2011), Michael et al. (2013), and Ahmed et al. (2015)). Gothwal and Shashidhar (2015) discussed the role that commonly water and wastewater treatment processes may play in the emergence, transport, and dissemination of antibiotic resistance in the environment. The state of the art in aqueous environmental analysis of antibiotics, focusing on sample preparation, analyte stability and degradation, and matrix effects were also reviewed by several authors (e.g. Díaz-Cruz and Barceló (2006) and Seifrtová et al. (2009)).

During last years, European Commission has increasingly promoted and supported several projects, actions, and initiatives to mitigate the widespread antimicrobial resistance, including surveillance of antibiotic consumption and research on environmental contamination by these drugs in Europe (EC, 2011; CDC, 2015; ECDC, 2015a; EFSA, 2015; EMA, 2015a; EC, 2015b,c). In this context, this review aims to provide an up to date comprehensive overview on the occurrence of antibiotics in the different aqueous environmental systems across the Europe, focusing in relating this topic to antibiotic consumption in human and veterinary medicine and their physicochemical properties and dynamic in the environment. Some issues which must be considered in the future for the progress of understanding related to the risks of the antibiotics in the aquatic environment and adjustment of the preventive measures are addressed. Moreover, the knowledge gaps in the currently available information on these subjects are emphatically identified and discussed along this literature review.

2. The global problem of antibiotics

2.1. The miracle under threat

The discovery of antibiotics is considered the greatest scientific and medical milestone of the 20th century. Their development and use in human and veterinary medicine resulted in the significant reduction of the mortality and morbidity rates of socially and epidemiologically significant infectious diseases such as tuberculosis, syphilis, pneumonia, gonorrhea, and communicable diseases of childhood. Consequently, the remarkable efficacy and efficiency of the antibiotics retains a sense of the miraculous (Alanis, 2005).

The modern antibiotic era traced back to researches performed by Paul Ehrlich and Alexander Fleming. In the beginning of the 90s, Ehrlich developed the systematic screening approach to concretize the idea of discovering magic bullets, drugs characterized by their selective toxicity for the infectious diseases treatment. These drug search strategy led to the discovery of sulfonamides, the first clinically successful broad spectrum antibiotics, by Bayer chemists in 1935 (Gradmann, 2011). On the other hand, it was the penicillin discovery by Fleming in 1929, and the subsequent developments in their purification by Howard Florey and Ernest Chain during World War II, that led the antibiotic revolution (Moore, 1999). In 1946, a year after the Nobel Prize in Medicine be awarded to Fleming, Florey, and Chain, penicillin started to be produced at industrial scale and commercialized in the open market. Thenceforward, hundreds of antibiotics have been developed for *anti*-infective therapy, beginning a new era for medicine, called the antibiotics era (Aminov, 2010; Schlipköter and Flahault, 2010). Therefore, this array of drugs have been saving the lives of millions of people and animals for >60 years. Nonetheless, the miracle of these special drugs has been increasingly threatened by the emergence, dissemination, and persistence of antibiotic resistance (Straand et al., 2008; WHO, 2014).

Antibiotic resistance is an adaptive genetic trait possessed or acquired by bacteria subpopulations, enabling them to survive and grow in the presence of the antibiotic agent at therapeutic concentration that would normally inhibit or destroy these microorganism. Despite

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