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Global review and analysis of erythromycin in the environment: Occurrence, bioaccumulation and antibiotic resistance hazards☆

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ABSTRACT

Environmental observations of antibiotics and other pharmaceuticals have received attention as indicators of an urbanizing global water cycle. When connections between environment and development of antibiotic resistance (ABR) are considered, it is increasingly important to understand the life cycle of antibiotics. Here we examined the global occurrence of erythromycin (ERY) in: 1. wastewater effluent, inland waters, drinking water, groundwater, and estuarine and coastal systems; 2. sewage sludge, biosolids and sediments; and 3. tissues of aquatic organisms. We then performed probabilistic environmental hazard assessments to identify probabilities of exceeding the predicted no-effect concentration (PNEC) of 1.0 $\mu g\,L^{-1}$ for promoting ABR, based on previous modeling of minimum inhibitory concentrations and minimal selective concentrations of ERY, and measured levels from different geographic regions. Marked differences were observed among geographic regions and matrices. For example, more information was available for water matrices (312 publications) than solids (97 publications). ERY has primarily been studied in Asia, North America and Europe with the majority of studies performed in China, USA, Spain and the United Kingdom. In surface waters 72.4% of the Asian studies have been performed in China, while 85.4% of the observations from North America were from the USA; Spain represented 41.9% of the European surface water studies. Remarkably, results from PEHAs indicated that the likelihood of exceeding the ERY PNEC for ABR in effluents was markedly high in Asia (33.3%) followed by Europe (20%) and North America (17.8%). Unfortunately, ERY occurrence data is comparatively limited in coastal and marine systems across large geographic regions including Southwest Asia, Eastern Europe, Africa, and Central and South America. Future studies are needed to understand risks of ERY and other antibiotics to human health and the environment, particularly in developing regions where waste management systems and treatment infrastructure are being implemented slower than access to and consumption of pharmaceuticals is occurring.

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1. Sources and hazards of erythromycin in the environment

Increasing population growth and expanding urban centers have strained water supplies, resulting in a new normal for water management (Postel, 2010) and an urban water cycle in many parts of the world. Such alterations and anthropogenic stressors in aquatic systems are further influenced by parallel increases in chemical access and consumption and concentration of chemical usage in urban regions. Unfortunately, wastewater treatment infrastructure and other waste management systems vary among geographic regions with ~80% of global sewage released as untreated waste to the environment (UNEP, 2015). Herein, traditional pollutants and contaminants of emerging concern, including pharmaceuticals, require attention to reduce risks to public health and the environment, particularly in developing countries (Hughes

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et al., 2013; Kookana et al., 2014). Specifically, among active pharmaceutical ingredients (APIs), antibiotics have become a major environmental and public health concern due to their capacity to alter microbial structure and functions and to influence development of antimicrobial resistance (Boxall et al., 2012; Singer et al., 2014).

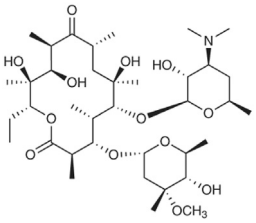
Though there are many classes of antibiotics, perhaps the most common is the macrolide class, which is commonly used for both human and veterinary medicine. Within the macrolides, erythromycin (ERY) was the first antibiotic clinically used to treat human infections (Gaynor and Mankin, 2003, Table 1). This pharmaceutical is considered a wide-spectrum antibiotic, acting against gram-positive and some gram-negative bacteria (Kanfer et al., 1998); it is commonly prescribed for lower and upper respiratory tract, and some soft-tissue, infections (McArdell et al., 2003). The mechanism of action of erythromycin is through binding to the 50S subunit of the bacterial ribosome to inhibit protein synthesis (Liu et al., 2014; Moffit, 1991). After administration, most of the compound is concentrated and metabolized in the liver and then excreted in the bile, but about 5% of the antibiotic is excreted in the active form (unchanged parent drug) through urine (Hardman et al., 2006).

ERY was first discovered in 1952 by McGuire and co-workers as metabolite product from a strain of *Streptomyces erythraeus*, which was later assigned to the genus *Saccharopolyspora*. This microorganism inhabits the soil, and thus EREY could be naturally produced by this bacterium in the environment. Before *Saccharopolyspora erythraea* had been used for industrial-scale production of ERY, Katz and Khosla (2007) argued that the erythromycin biosynthesis by this bacterium helped the survival against predators that inhabit soil systems. However, the vast majority of ERY residues in environmental matrices result from anthropogenic sources (Liu and Wong, 2013). Specifically, introduction of ERY to the environment primarily begins via excretion in feces and urine of humans and animals. Though most of ERY and other antibiotics residues are excreted into the sewage system reaching wastewater treatment plants (WWTPs) (Kümmerer, 2009), improper disposal of unused medicines through sewage also reaches WWTPs (Glassmeyer et al., 2009; Persson et al., 2009). ERY was found to be fairly resistant to different levels of treatment in WWTP facilities from south China with ubiquitous occurrence in final effluent (Leung et al., 2012; Xu et al., 2007). Therefore, WWTP effluents are considered one of main contributors to the spread ERY and other antibiotics in the environment (Boxall et al., 2012; Carvalho and Santos et al., 2016; Michael et al., 2013). Additionally, effluents from pharmaceutical production facilities and hospitals can be considerable point sources of ERY (Qarni et al., 2016; Santos et al., 2013). In fact, most pharmaceuticals are manufactured in lower income countries, and shifting of global manufacturing to Asia and other underdeveloped regions inherently influences environmental exposure patterns of APIs in these countries (Kookana et al., 2014).

During wastewater treatment, ERY can bind to biosolids and contaminate sludge. Because application of sewage sludge as a fertilizer on agricultural land represents a common practice, ERY residues can be transported through soil and will ultimately reach groundwater (Radjenović et al., 2009a; Yan et al., 2014). Waste from veterinary applications also results in ERY introductions to the environment (Hou et al., 2015; Sim et al., 2011). For example, transport of ERY from soil to groundwater also occurs when manure is also used as an agricultural fertilizer (Sui et al., 2015; Watanabe et al., 2010). Further, aquaculture effluent presents another pathway of antibiotics to the environment (Bottoni et al., 2010). For example, ERY is often used to treat nocardiosis caused by the pathogen *Nocardia seriolae*, in addition to the bacterial kidney disease and lactococcosis caused by the bacterial pathogens *Renibacterium salmoninarum* and *Lactococcus garvieae*, respectively (Moffitt and Kiryu, 1999; Vendrell et al., 2012; Wang et al., 2009; Xia et al., 2015).

Despite increasing public health and environmental studies, no legal limits exist for ERY in the environment. In United States of

Table 1
Physicochemical profile of erythromycin.

CASRN	114-07-8
Molecular structure	
Molecular formula	$C_{37}H_{67}NO_{13}$
Molecular weight	733.937
Dissociation constant, pKa	8.89
Octanol/Water Partition Coefficient (log k_{ow})	3.06
Bioconcentration factor (BCF)	9.28–142.6[a]; 49[b]
Solubility	4.2–2000 mg/L at 25 °C in water and greater solubility in acetone, ethyl ether, ethanol and chloroform
Henry Law constant	5.42×10^{-29} atm-m^3/mol at 25 °C (est)
Melting point	191 °C
Vapor pressure	2.12 × 10–25 mm Hg at 25 °C
Color and form	White or slightly yellow crystals or powder; hydrated crystals in water
Odor	Odorless
Taste	Bitter
Half-life, days* and hours**	11.5* (sediments)[c], 20* (soil), 5.8–365* (pond water system)[d], 1.6–2.0** (humans)[e]

[a] Range reported in the U.S. Environmental Protection Agency's EPI Suite program, a physical/chemical property and environmental fate estimation program (U.S. Environmental Protection Agency for Estimation Programs Interface - EPI Suite v4.11, November 2012).
[b] Value reported in the US National Institutes of Health's data bank (US National Library of Medicine - National Center for Biotechnology Information).
[c] McArdell et al. (2003).
[d] Jessick et al. (2013).
[e] Hardman et al. (2006).

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