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Development of a semi-quantitative risk assessment model for evaluating environmental threat posed by the three first EU watch-list pharmaceuticals to urban wastewater treatment plants: An Irish case study

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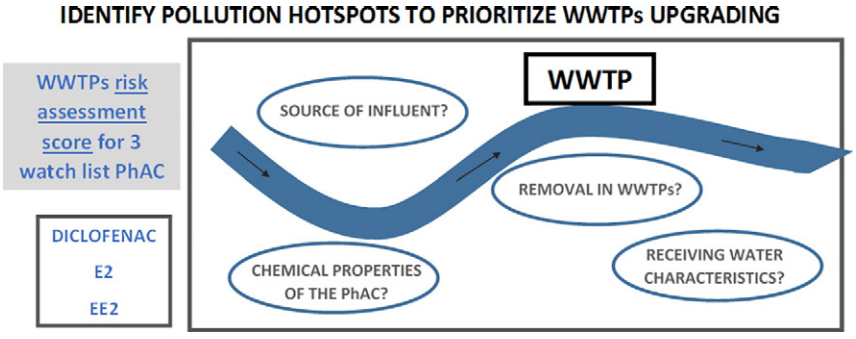
HIGHLIGHTS

- Lack of data regarding the likelihood of one given pharmaceutical to be found in river
- Semi-quantitative risk assessment (RA) proposed for three watch-list PhACs
- The RA is based on influent source, PhACs properties, WWTP removal and receiving water
- A method to identify pollution hotspots and WWTPs responsible for PhACs release is provided

GRAPHICAL ABSTRACT



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ABSTRACT

Contamination of receiving waters with pharmaceutical compounds is of pressing concern. This constitutes the first study to report on the development of a semi-quantitative risk assessment (RA) model for evaluating the environmental threat posed by three EU watch list pharmaceutical compounds namely, diclofenac, 17-beta-estradiol and 17-alpha-ethinylestradiol, to aquatic ecosystems using Irish data as a case study. This RA model adopts the Irish Environmental Protection Agency Source-Pathway-Receptor concept to define relevant parameters for calculating low, medium or high risk score for each agglomeration of wastewater treatment plant (WWTP), which include catchment, treatments, operational and management factors. This RA model may potentially be used on a national scale to (i) identify WWTPs that pose a particular risk as regards releasing disproportionally high levels of these pharmaceutical compounds, and (ii) help identify priority locations for introducing or upgrading control measures (e.g. tertiary treatment, source reduction). To assess risks for these substances of emerging concern, the model was applied to 16 urban WWTPs located in different regions in Ireland that were scored for the three different compounds and ranked as low, medium or high risk. As a validation proxy, this case study used limited monitoring data recorded at some these plants receiving waters. It is envisaged that this semi-quantitative RA approach may aid other EU countries investigate and screen for potential

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risks where limited measured or predicted environmental pollutant concentrations and/or hydrological data are available. This model is semi-quantitative, as other factors such as influence of climate change and drug usage or prescription data will need to be considered in a future point for estimating and predicting risks.

1. Introduction

Pollution of European receiving waters containing pharmaceuticals is a ubiquitous phenomenon (Barbosa et al., 2016; Verlicchi and Zambello, 2016; Tiedeken et al., 2017). Pharmaceuticals are a class of emerging environmental contaminants that are widely used in human and veterinary medicine and are essential to modern healthcare (Streck, 2009; Kosma et al., 2014). From here on, these compounds will be referred to as pharmaceutically active chemicals (PhACs). Nevertheless, there are growing concerns about the negative impacts that may result from continuous contamination of the environment with PhACs. This research is important because of the potential toxic effects for aquatic biota and human health that may result from chronic exposure to PhACs (Miege et al., 2008; Streck, 2009; Kosma et al., 2014). PhACs exhibit wide variation in function, chemical structure and physiochemical properties, making it difficult to generalize about their behaviour, persistence or impact in the environment. PhACs are also designed to be biologically active, have a specific mode of action and to be persistent in the body, meaning they can impact humans and wildlife at trace concentrations that are often hard to detect and quantify using traditional analytical methods (Kosma et al., 2014). A large number of PhACs have been detected in wastewater treatment plants (WWTPs) influents and effluents and surface, ground and drinking water worldwide in recent years (Nikolaou et al., 2007; Cirja et al., 2008; Streck, 2009; Verlicchi and Zambello, 2016). The impacts of chronic exposure to trace concentrations of many PhACs on wildlife and human health may be severe (Verlicchi et al., 2012b), thus it is critical to limit as much as possible the concentrations of this class of contaminants in aquatic environments.

Until recently, environmental regulations worldwide had not required explicit testing for any PhACs in water bodies. However, given the growing concern about contamination of the aquatic environment with these compounds, legislation has recently begun to acknowledge this potential problem. The Water Framework Directive (WFD) requires that all EU member states prepare river basin management plans (RBMPs) to address the many issues relating to water quality and protection in a holistic manner. In response to growing EU concern about the release of untreated PhACs into the aquatic environment, three compounds were included on in the first EU watch list in 2013: diclofenac (CAS# 15307–79-6, hereafter referred as DCL), 17-beta-estradiol (CAS# 50–28 − 2, hereafter referred as E2) and 17-alpha-ethinylestradiol (CAS# 57–63-6, hereafter referred as EE2). E2 and EE2 can impact the endocrine system of humans or wildlife where there is growing fears that chronic exposure to these endocrine disrupting chemicals or endocrine disrupting chemicals (EDCs) even at low concentrations (ng/L) (in bathing or drinking water, for example) may be linked to adverse human health conditions (Hernando et al., 2006; Streck, 2009). Similar to PhACs as a whole, EDCs are generally thought to be transported into the aquatic environment mostly via incomplete removal at wastewater treatment plants (WWTPs) (Streck, 2009). It is relevant to note that the European Commission implemented decision 495 of 20 March 2015 that expanded substances or groups of substances on the watch list to 10 in the field of water policy (Barbosa et al., 2016). This review focuses solely on the first three substances DCL, E2 and EE2 as there is a requirement to investigate policy implications for Ireland of these PhACs in receiving waters in the first instance.

Tiedeken et al. (2017) conducted a 20 year systematic review of 3945 potentially relevant articles published between 1995 and 2015 that produced a EU-wide database consisting of 1268 publications on DCL, E2 and EE2 in receiving waters. Overall, European surface water concentrations of DCL are typically reported below the proposed annual average environmental quality standard (AA EQS) of 100 ng/L with only a few extreme values exceeding this threshold (up to 1200 ng/L according to Rivera-Utrilla et al. (2013)). E2 and EE2 surface water concentrations are typically below 50 ng/L and 10 ng/L respectively, but these concentrations greatly exceed the proposed AA EQS values for these compounds (0.4 and 0.035 ng/L respectively). Furthermore, levels of these PhACs are frequently reported to be disproportionately high in EU receiving waters (up to 200 ng/L and 831 ng/L respectively in surface water according to Tiedeken et al. (2017)), particularly in effluents at control points that require urgent attention. Furthermore, the number of articles produced by each of the 28 EU countries along with Switzerland, Norway and Turkey varied greatly on sources, monitoring or control measures for DCL, E2 and EE2 over this 20-year systematic review period, where Spain, Germany and the United Kingdom contributed 707 (56%) of all reports. However, 24 and 16 EU countries produced under 50 and 20 reports respectively on these PhACs in their national receiving waters; consequently, very few countries have reported on use predicted or measured environmental concentrations to underpin modelling or to inform risks in their river basins (ter Laak et al., 2010; Guillén et al., 2012). Overall, it was found that DCL and EE2 enter European aquatic environment mainly following human consumption and excretion of therapeutic drugs, and by incomplete removal from influent at urban WWTPs. E2 is a natural hormone excreted by humans, which also experiences incomplete removal during WWTP treatments. Thus, WWTPs (initially not designed to remove PhACs) are considered as pressure point for control of PhACs in aquatic environment (Tiedeken et al., 2017). Current conventional analytical methods (i.e. LC-MS/MS) are sufficiently sensitive for the detection and quantification of DCL, but generally not for E2 and EE2 (at the very low EQS levels proposed for those compounds, i.e. 0.4 and 0.035 ng/L respectively, levels that cannot be reached for most of the standards chemical analysis labs) (Streck, 2009), thus alternative, ultra-trace, time-integrated monitoring techniques such as passive sampling are needed to inform water quality for these estrogens (Buchberger, 2011; Wille et al., 2012). Another emerging potential solution to the problem of low EQS values of E2 and EE2 is the use of biological effects monitoring techniques (Streck, 2009; Kunz et al., 2015; Simon et al., 2015). WWTPs are today widely considered as the main vector of PhACs through the aquatic environment (Hernando et al., 2006; Tiedeken et al., 2017). However, in Ireland as well in numerous other EU countries the number of reliable WWTPs and surface/ground waters monitoring data is still limited (Tiedeken et al., 2017) making difficult the direct and quantitative risk analysis associated to the selected PhACs. The aforementioned limitations of reliable data at WWTPs makes it difficult to directly quantify risks for aquatic environment. As an alternative, the application of a risk assessment (RA) method appeared to be the best way to obtain a full picture of the potential contamination of surface waters by the three selected PhACs and to identify PhACs emission hotspots. Other RA models related to PhACs in the aquatic environment aiming at: (i) prioritize certain compounds (Guillén et al., 2012), (ii) the evaluation of the ecological/environmental risk related to the presence of PhACs in rivers downstream from WWTP (e.g. Hernando et al., 2006; Ginebreda et al., 2010; Gros et al., 2010; Kosma et al., 2014; Pereira et al., 2015), (iii) the evaluation of PhACs concentrations in surface water based on their physico-chemical characteristics (Lindim et al., 2017), or on their