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Active pharmaceutical ingredients entering the aquatic environment from wastewater treatment works: A cause for concern?

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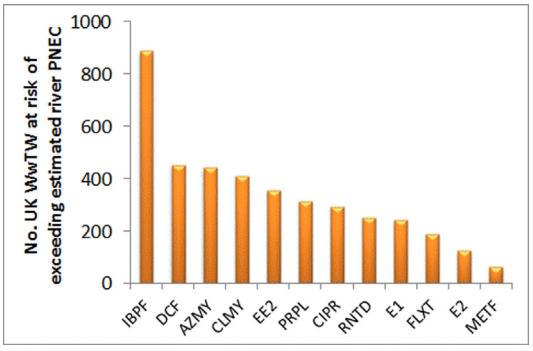
HIGHLIGHTS

- New data for 19 active pharmaceuticals and 4 metabolites in 45 UK sewage works
- Detailed data analysis provided for removal efficiency for the pharmaceuticals
- First time risk is related to measured concentrations for all UK sewage works.
- Up to 890 works may be causing exceedances of downstream water estimated PNECs.
- Ibuprofen, diclofenac, ethinyloestradiol and 2 antibiotics of greatest concern

GRAPHICAL ABSTRACT



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ABSTRACT

This work reports on the variation in wastewater treatment works (WwTW) influent concentrations of a wide variety of active pharmaceutical ingredients (APIs), their removal efficiency, effluent concentrations and potential risks to the aquatic environment. The research is based on data generated from two large UK-wide WwTW monitoring programmes. Taking account of removal of parent compound from the aqueous phase during treatment in combination with estimates of dilution available it is possible to prioritise the APIs of greatest risk of exceeding estimates of predicted no effect concentrations (PNEC) in receiving waters for all WwTW in the UK. The majority of substances studied were removed to a high degree, although with significant variation, both within and between WwTW. Poorer removal (between influent and effluent) was observed for ethinyloestradiol, diclofenac, propranolol, the macrolide antibiotics, fluoxetine, tamoxifen and carbamazepine. All except the last two of these substances were present in effluents at concentrations higher than their respective estimated PNEC (based on measurement of effluents from 45 WwTW on 20 occasions). Based on available dilution data as many as 890 WwTW in the UK (approximately 13% of all WwTW) may cause exceedances of estimated riverine PNECs after mixing of their effluents with receiving waters. The overall degree of risk is driven by the toxicity value selected, which in itself is controlled by the availability of reliable and relevant ecotoxicological data and consequently the safety factors applied. The dataset and discussion, provides information to assist in the future management of these types of chemicals.

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1. Introduction

The use and environmental prevalence of pharmaceuticals increases on an annual basis due to a variety of reasons including the widening array of medical treatments available, greater availability of medicines across the world, affordability, population growth, population ageing (in some countries) and changing perspectives towards, for example, pain (Jelic et al., 2011). Active Pharmaceutical Ingredients (API) are detected throughout the environment in water, soil, sediment, sludge as well as in drinking waters in some countries (Kasprzyk-Hordern et al., 2008; Zorita et al., 2009; Wahlberg et al., 2011; Jones et al., 2014; Lees et al., 2016). Although the mere presence of pharmaceutical is not always associated with harm to the environment or human health, concerns are rising associated with antimicrobial resistance and chronic impacts on biodiversity including endocrine disrupting effects on fish (Levado et al., 2004; Jobling et al., 2005; Tyler et al., 2008). The main source of occurrence of APIs in the river environment is from human use of pharmaceuticals, via the continuous discharge of effluent from the Wastewater Treatment Works (WwTW) (Gardner et al., 2012; Melvin and Leusch, 2016). Hence, investigating the occurrence, fate and risk of APIs is currently of great interest to regulators and the water industry alike, with a focus to better understand the loadings entering WwTW and the observed within and between works variation in removal efficiencies and concentrations often observed for APIs (Gardner et al., 2013).

The range of concentrations found for pharmaceuticals studied in the UK is similar to that observed in continental Europe as well as in the USA (Kolpin et al., 2002; Ashton et al., 2004; Hope et al., 2012; Bradley et al., 2016; Burns et al., 2017). Table 1 provides examples of other reported data for APIs determined as part of this research, rather than a complete list of all APIs detected in effluent and receiving waters. Other studies have also shown that there is a clear association between the number of pharmaceuticals used in a society and the levels of API found in receiving water bodies ranging from API concentration of typically <100 ng/l in the surface and groundwater and below 50 ng/l in treated drinking water (WHO, 2011; Furlong et al., 2017) to higher levels reported adjacent to production facilities (Phillips et al., 2010). Predicted no effect concentrations (PNECs) have been reported for some APIs below 1 ng/l and APIs such as diclofenac (CAS 15307-79-6), 17-beta-estradiol (E2) (CAS 50-28-2) and 17-alpha-ethinylestradiol (EE2) (CAS 57-63-6) are on the European Water Framework Directive (WFD) 'watch list' (EU, 2013). This requires member states to gather monitoring data in order to assess risk to the environment, leading to significant sources of APIs needing to be quantified and factors controlling the discharge of APIs carefully considered along with impacts on receiving water ecology, including effects of mixtures (Bound and Voulvoulis, 2006).

Many countries have therefore started monitoring programs to investigate the exposure of APIs in order to gain a better understanding of their sources, fate and risk (Falås et al., 2012). The Chemical Investigation Program (CIP) in the UK is a large ongoing investment being undertaken by the water industry to assist the UK in meeting its obligations under the WFD to monitor concentrations of priority chemicals including APIs in WwTW influent, intermediate processes and effluent as well as assessing their risk to receiving waters (Gardner et al., 2013). The first phase of the CIP (named CIP1 here) was a project that ran from 2012 to 2015 with one of its aims to investigate the fate of trace substances (including 11 APIs) in influent, effluent and intermediate WwTW

Table 1
Average aquatic concentrations for APIs of interest to this research found in river water, as well as usage, excretion and removal in WwTW.

API	Therapeutic class	Upstream (μg/l)	Influent (μg/l)	Effluent (μg/l)	WwTW removal (%)	Down stream (μg/l)	UK consumption (ton/year), 2009 and 2011	Excreted unchanged compound (%)
Aspirin (acetylsalicylic acid)	Anti-inflammatory/analgesics	NA	NA	NA	NA	<0.0005[b]	130[d]	<1[b]
Atenolol	Beta blocker	NA	NA	NA	NA	0–0.56[b]	28[e]	90[f]
Azithromycin	Antibiotic	NA	0.163[l]	0.030[l]	90[l]	NA	NA	NA
Carbamazepine	Antiepileptic	NA	2.593[b]	3.117[b]	ND[b]	0.0005–0.356[b]	48[e]	3[b]
Ciprofloxacin	Antibiotic	NA	1.090[l]	0.052[l]	97[l]	NA	NA	NA
Clarithromycin	Antibiotic	NA	0.524[l]	0.092[l]	91[l]	NA	NA	NA
Diclofenac	Anti-inflammatory	<0.020[a]	0.107–0.981[c]	0.599[a]	70–92[c]	0.154[a]	28[e]	15[f]
Erythromycin	Antibiotic	<0.010[a]	2.0[k]	0.109[a]	25–91[i]	0.159[a]	3[d]	25[f]
Oestrogen (E1)	Natural hormone	NA	0.042[g]	0.011–0.025[g]	58–96[g]	NA	NA	NA
Oestradiol (E2)	Contraceptive	NA	0.016[g]	0.0013–0.0039[g]	89–96[g]	NA	NA	NA
Ethinylestradiol (EE2)	Contraceptive	NA	0.0017[g]	0.00033–0.00078[g]	53–71[g]	NA	NA	NA
Fluoxetine	Psychiatric drugs	NA	0.070[k]	0.023[j]	33–100[h]	NA	6.4[m]	NA
Ibuprofen	Analgesic	0.432[a]	14.0[k]	4.201[a]	90–100[i]	1.105[a]	258[e]	10[f]
Oxytetracycline	Antibiotic	NA	1.09[l]	0.029[l]	99[l]	NA	NA	NA
Ofloxacin	Antibiotic	NA	0.081[l]	0.023[l]	89[l]	NA	NA	NA
Propranolol	Antihypertensive	0.010[a]	0.542[b]	0.093[a] 0.388[b]	28[b]	0.041[a]	15[e]	<0.5[b]
Tamoxifen	Anti-cancer	<0.010[a]	0.0002–0.015[c]	<0.010[a]	32–45[c]	<0.010[a]	NA	NA

ND = not detected; NA = not available.
[a] Ashton et al. (2004).
[b] Kasprzyk-Hordern et al. (2008).
[c] Zhou et al. (2009).
[d] 2006 sales data for Wales; Kasprzyk-Hordern et al. (2008).
[e] IMS figure on active ingredient sales; IMS (2016).
[f] WHO (2011).
[g] Heffley et al. (2014).
[h] Clara et al. (2005).
[i] Li et al. (2015).
[j] Gardner et al. (2012).
[k] Gardner et al. (2013).
[l] Singer et al. (2014).
[m] Boxall et al. (2014).