Environmental Pollution 220 (2017) 1447–1455

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Widespread, routine occurrence of pharmaceuticals in sewage effluent, combined sewer overflows and receiving waters☆

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ARTICLE INFO

Article history:
Received 11 August 2016
Received in revised form
27 October 2016
Accepted 28 October 2016
Available online 6 November 2016

Keywords:
Pharmaceuticals
Water quality
Effluent
Pollution
Emerging contaminants

ABSTRACT

Research addressing the occurrence, fate and effects of pharmaceuticals in the aquatic environment has expanded rapidly over the past two decades, primarily due to the development of improved chemical analysis methods. Significant research gaps still remain, however, including a lack of longer term, repeated monitoring of rivers, determination of temporal and spatial changes in pharmaceutical concentrations, and inputs from sources other than wastewater treatment plants (WWTPs), such as combined sewer overflows (CSOs). In addressing these gaps it was found that the five pharmaceuticals studied were routinely (51–94% of the time) present in effluents and receiving waters at concentrations ranging from single ng to μg L^{-1}. Mean concentrations were in the tens to hundreds ng L^{-1} range and CSOs appear to be a significant source of pharmaceuticals to water courses in addition to WWTPs. Receiving water concentrations varied throughout the day although there were no pronounced peaks at particular times. Similarly, concentrations varied throughout the year although no consistent patterns were observed. No dissipation of the study compounds was found over a 5 km length of river despite no other known inputs to the river. In conclusion, pharmaceuticals are routinely present in semi-rural and urban rivers and require management alongside more traditional pollutants.

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1. Introduction

An increasing global population is placing great strain on over 65% of the Earth's rivers with chemical pollution one of the main causes of degradation and biodiversity loss in aquatic ecosystems (Vorosmarty et al., 2010). In chemical pollution research there has been an increasing focus on emerging contaminants over recent decades (Daughton and Ternes, 1999) which enter the aquatic environment following excretion or disposal to the sewer system and passage through wastewater treatment plants (WWTPs) (Kolpin et al., 2002). It is now widely considered that WWTP effluent is the dominant route by which pharmaceuticals enter the aquatic environment (Heberer, 2002; Daughton, 2004; Jones et al., 2005; Tambosi et al., 2010). Although the likelihood of human health impacts due to pharmaceuticals in the environment is low their presence in continually discharged effluent is a major ecological concern due to the potential for effects on aquatic organisms at trace concentrations (Daughton, 2001; Cleuvers, 2003, 2004; Fent et al., 2006; Caliman and Gavrilescu, 2009; Kümmerer, 2009; Santos et al., 2010).

Research into pharmaceutical pollution is expanding largely due to increased concern over potential adverse effects and advancements in the analytical techniques necessary to detect such compounds at trace concentrations (Daughton, 2001; Williams, 2005). Although chemical analysis methods have been improved greatly, there remains a dearth of research which uses these to quantify the occurrence of pharmaceuticals throughout river catchments over periods of time, as has been done for other chemicals such as metals, nutrients (Neal et al., 2012) and pesticides (Bundschuh et al., 2014). Very little work has been conducted in large parts of Africa, Asia, the Middle East and South America and even within those countries with a relatively high level of research the number of studies undertaken remains very small compared to other groups of chemicals (Hughes et al., 2013). Of 155 published pharmaceutical monitoring studies 80% were reported to have been carried out in the US and Europe (Hughes et al., 2013). Furthermore,

☆ This paper has been recommended for acceptance by Dr. Chen Da.

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http://dx.doi.org/10.1016/j.envpol.2016.10.087
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where research has been undertaken, because only a limited number of studies exist, spatial bias exists in pharmaceutical occurrence datasets. For example, UK pharmaceutical pollution monitoring is heavily clustered around south east England and parts of south Wales with very few studies in central, western and northern England or Scotland where large urban areas exist. Where studies are present they often rely on non-repeated sampling and have typically provided very few details on the adopted sampling regime, making it difficult to draw conclusions about the reliability or representativeness of the data presented (Hughes et al., 2013).

In addition to WWTPs, combined sewer overflows (CSOs) and misconnections to storm water drains, which could lead to the discharge of untreated sewage effluent to receiving waters, have been identified as potential sources of pharmaceutical pollution. Despite this, there are very few studies which attempt to examine the contribution they make to overall pharmaceutical loads in rivers (Boyd et al., 2004; Kolpin et al., 2004). This is of concern as it has been hypothesised that such non-WWTP point sources may actually be the key contributor of high pharmaceutical concentrations in reaches far from WWTP effluent outfalls and where dissipation has not been found to occur downstream of WWTPs (Ellis, 2006). Residual low levels of pharmaceuticals have been detected tens or hundreds of kilometres downstream of WWTP outfalls (Waiser et al., 2011), demonstrating the potential for widespread, catchment-level impacts. This presents a pressing research need given the assumption in many risk assessment models of first-order, in-stream decay of pharmaceuticals (Schowanek et al., 2001). Only 16% of monitoring studies included in a recent critical review paper collected samples more than 1 km downstream of WWTP outfalls, indicating a tendency for research to focus on the effluent dominated reaches immediately downstream (Hughes et al., 2013). This is understandable given the likelihood that these areas are most affected by pharmaceutical pollution but this often leaves long reaches of catchments with little or no pharmaceutical monitoring data available.

Research examining temporal variation in pharmaceutical concentrations in receiving waters is also rare, despite some evidence demonstrating high degrees of variation over hourly and daily periods (Kanda et al., 2003) as well as seasons (Lindholm-Lehto et al., 2016; Papageorgiou et al., 2016). Given the tendency towards non-repeated grab sampling of receiving waters it is unlikely that such variation has so far been adequately captured in existing monitoring datasets and they may therefore currently give an inaccurate description of the occurrence of pharmaceuticals in rivers (Ort and Gujer, 2006; Ort et al., 2010a, b).

Given the highlighted research gaps, the aims of the current study were to: carry out repeated sampling of river reaches throughout an eighteen month period for five pharmaceuticals; monitor the chemicals' presence in WWTP and CSO effluent as well as their receiving waters; undertake diurnal monitoring of pharmaceuticals in the receiving waters of WWTPs; and examine dissipation of the study compounds over a 5 km river reach.

2. Methods

2.1. Study area and sampling sites

The Aire and Calder catchments, West Yorkshire, UK, are ideal for studying the occurrence of pharmaceuticals in rivers given the 105 WWTPs that discharge effluent into them (Fig. 1). The catchments are heavily urbanised in the lower reaches with the West Yorkshire Urban Area being one of the ten most populous areas of the UK and being home to around 1.5 million people (Pointer, 2005). There are also a number of smaller towns and villages in the semi-rural and rural upland parts of the catchments. In addition to the WWTPs there are estimated to be 70 CSOs spread across the entire catchment area (Environment Agency, 2010). The total catchment areas of the Aire and Calder above the tidal limit are 1932 and 899 km^2 respectively. Mean annual discharges in the downstream reaches of the catchments are 36 and 19 $m^3\ s^{-1}$ (Carter et al., 2006). Seven WWTPs (supplementary material S1 for treatment techniques and populations served) were monitored monthly for eighteen months and five CSOs were sampled during periods of intensive rainfall which caused them to discharge. More spatially intensive reach monitoring, below one of the WWTPs (Knostrop), was undertaken on seven occasions to look at pharmaceutical dissipation downstream of specific WWTP discharges. This was done over a 5 km length of river; the distance to the next WWTW downstream where more effluent would have entered the river. Diurnal sampling was undertaken on two occasions at Garforth and Oulton WWTPs with samples being collected every 3 h at each.

2.2. Sample collection procedure

Grab samples of effluent (WWTP and CSO) and receiving channel water (0.8 L) for all field surveys were collected in 1 L amber silanised glass bottles with Teflon® lined caps (Fisher Scientific, Leicestershire, UK) and kept chilled in the dark during transit. Samples of WWTP effluent and receiving waters were collected at the start of each month at the same time of day to minimise errors associated with diurnal fluctuations in pharmaceutical concentrations (Kanda et al., 2003). Receiving water samples were collected at a point of five times the stream width downstream of the effluent outfall to allow for mixing (Morris, 2013). Samples were collected from the centre of the stream at 50% depth in-line with established guidelines (USGS, 2006) where possible, or otherwise, due to the size of the channel and bank topography, at the bankside (downstream of Heaton Lodge, Horbury Junction, Oulton/Lemonroyd, and Knostrop WWTP). CSO samples were collected during storm events when the CSOs were discharging to streams and the release of untreated effluent could have an impact on them. All apparatus and glassware used during sample collection and preparation was thoroughly washed with 100% methanol (1 x) and de-ionised water (3 x) prior to each use to remove potential contamination. On return to the laboratory, samples were stored in the dark at 4 °C and extracted within 48 h.

2.3. Study compounds

Five study compounds (Table 1) were chosen based on risk quotients (RQ) (ratio of predicted or maximum environmental concentration to predicted no effect concentration) produced in previously published studies (Jones et al., 2002; Thompson, 2006; Yamamoto et al., 2009). A RQ $\geq$ 1 indicates the potential for impacts on aquatic organisms so this was used as the basis for selection and the number of compounds limited to five to focus on high risk substances.

2.4. Analysis of receiving water and effluent samples

Pharmaceutical standards were used to create working and stock solutions in dilution series for calibration of analytical instruments. All pharmaceuticals were supplied by Sigma-Aldrich Company Ltd. (Dorset, UK) and were of the highest purity available (>99%). Individual stock standard solutions (1000 ng L^{-1}) were prepared on a weight basis in 100% methanol and stored in the dark at −20 °C until used. A fresh working mixture solution of all pharmaceuticals was prepared by appropriate dilution of the individual stocks in methanol-water (20:80, v/v) immediately before

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