Environmental Pollution 220 (2017) 919–926

Contents lists available at ScienceDirect

Environmental Pollution

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

Linking changes in antibiotic effluent concentrations to flow, removal and consumption in four different UK sewage treatment plants over four years☆

Andrew C. Johnson [a, *], Monika D. Jürgens [a], Norihide Nakada [b], Seiya Hanamoto [b], Andrew C. Singer [a], Hiroaki Tanaka [b]

[a] NERC Centre for Ecology and Hydrology, Wallingford, Oxfordshire, OX10 8BB, United Kingdom
[b] Research Centre for Environmental Quality Management, Kyoto University, 1-2 Yumihama, Otsu, Shiga 520-0811, Japan

ARTICLE INFO

Article history:
Received 30 August 2016
Received in revised form
19 October 2016
Accepted 26 October 2016
Available online 10 November 2016

Keywords:
Antibiotics
Sulfamethoxazole
Clarithromycin
Trimethoprim
Tetracycline
Effluent

ABSTRACT

The arrival and discharge of seven antibiotics were monitored at two trickling filter sewage treatment plants of 6000 and 11,000 population equivalents (PE) and two activated sludge plants of 33,000 and 162,000 PE in Southern England. The investigation consisted of 24 h composite samples taken on two separate days every summer from 2012 to 2015 and in the winter of 2015 (January) from influent and effluent. The average influent concentrations generally matched predictions based on England-wide prescription data for trimethoprim, sulfamethoxazole, azithromycin, oxytetracycline and levofloxacin (within 3-fold), but were 3–10 times less for clarithromycin, whilst tetracycline influent concentrations were 5–17 times greater than expected. Over the four years, effluent concentrations at a single sewage plant varied by up to 16-fold for clarithromycin, 10-fold for levofloxacin and sulfamethoxazole, 7-fold for oxytetracycline, 6-fold for tetracycline, 4-fold for azithromycin and 3-fold for trimethoprim. The study attempted to identify the principal reasons for this variation in effluent concentration. By measuring carbamazepine and using it as a conservative indicator of transport through the treatment process, it was found that flow and hence concentration could alter by up to 5-fold. Measuring influent and effluent concentrations allowed assessments to be made of removal efficiency. In the two activated sludge plants, antibiotic removal rates were similar for the tested antibiotics but could vary by several-fold at the trickling filter plants. However, for clarithromycin and levofloxacin the variations in effluent concentration were above that which could be explained by either flow and/or removal alone so here year on year changes in consumption are likely to have played a role.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Prioritisation exercises for pharmaceuticals in the environment typically list antibiotics as one of the groups of highest concern (Besse and Garric, 2008; Christensen et al., 2009; Cooper et al., 2008). Their high ranking is linked not only to their high levels of consumption and toxicity to aquatic wildlife but also to concerns over possible links to antibiotic resistance which could have consequences for mankind (Ågerstrand et al., 2015). This was prompted by the co-occurrence of antibiotics and antibiotic resistance genes in some river environments (Amos et al., 2015; Huerta et al., 2013; Marti et al., 2013, 2014; Rodriguez-Mozaz et al., 2015). Geographic based modelling has been used as part of the risk assessment process for antibiotics (Johnson et al., 2015; Singer et al., 2014). If we should wish to remove more antibiotics in sewage treatment, it will be important to identify the factors associated with good performance. Currently this is difficult due to the surprisingly wide variety in effluent concentrations and apparent removal rates of similar antibiotics found in the literature.

There have been a number of studies which have examined temporal changes in antibiotic loadings in sewage treatment plants (STPs) ranging from daily (Coutu et al., 2013; Singer et al., 2014) to

Abbreviations: TRIM, trimethoprim; SMX, sulfamethoxazole; CBZ, Carbamazepine; CLAR, clarithromycin; AZO, azithromycin; TET, tetracycline; OXY, oxytetracycline; LEVO, levofloxacin; STP, sewage treatment plant; AS, activated sludge; TF, trickling filter.

☆ This paper has been recommended for acceptance by Klaus Kummerer.

* Corresponding author.
E-mail address: ajo@ceh.ac.uk (A.C. Johnson).

http://dx.doi.org/10.1016/j.envpol.2016.10.077
0269-7491/© 2016 Elsevier Ltd. All rights reserved.

seasonal (Gracia-Lor et al., 2012). Probably the most notable observation has been a seasonal increase in consumption for some antibiotics associated with winter. For example, in the Czech Republic and Switzerland, both clarithromycin and trimethoprim consumption doubled in winter (Coutu et al., 2013; Golovko et al., 2014; McArdell et al., 2003). For sulfamethoxazole, concentrations in influent rose by one-quarter to one-third in winter in the Czech Republic, Greece and China (Golovko et al., 2014; Kosma et al., 2014; Zhang et al., 2015), but in Portugal consumption was higher in spring than summer for the two antibiotics studied (azithromycin and ciprofloxacin) (Pereira et al., 2015). Sometimes the observed loadings (consumption) appear to be a lot higher than might have been expected from reviewing national or regional prescription data (Singer et al., 2014).

There have not been many studies reviewing the performance of different sewage treatment types with respect to antibiotics, but in a Chinese study a membrane bioreactor gave better removal performance for trimethoprim than a conventional activated sludge (AS) plant (Sui et al., 2011). In the UK there did not appear to be a consistent trend in performance between activated sludge plants or trickling filters with respect to oxytetracycline removal (Gardner et al., 2013).

Regarding seasonal changes in sewage treatment removal performance, for the Czech Republic it appeared that clarithromycin and trimethoprim removal improved by 20% in summer compared to winter, whilst levofloxacin and sulfamethoxazole improved by 10% (Golovko et al., 2014). In a Chinese study, trimethoprim removal performance improved from 30 to 80% in the Beijing summer, which might have been temperature related (Sui et al., 2011). In Portugal, higher azithromycin and ciprofloxacin removal was reported in summer compared to spring (80% versus 50%).

These studies imply consumption for some antibiotics could increase between two and four-fold in winter and removal performance decline by more than 2/3. However, this may not always lead to higher antibiotic concentrations in the river. Many Western countries experience their highest river flows in winter, so that dilution might rise 30-fold (Johnson, 2010) which more than compensates for relatively minor changes in seasonal antibiotic use or removal efficiency.

Changes in effluent concentrations of antibiotics over several years at the same STP has not been examined before. Nor have there been serious attempts to disentangle the reasons for variations in effluent concentrations over long time-scales. A better understanding of why there are differences in antibiotic discharge between STPs would assist both risk assessment and a strategy to improve their removal from wastewater.

Following the development of a method, which permitted the simultaneous analysis of several pharmaceuticals in wastewater including some key antibiotics, a study was prepared to look at several antibiotics in use in the UK. Sulfamethoxazole (SMX) is used to treat infections such as urinary tract, inner ear, prostitis and bronchitis. It is one of the sulphonamide group which entered the market in the 1970s and inhibits an enzyme involved in the synthesis of tetrahydrafolic acid (part of the thymidine metabolic pathway in DNA synthesis) (Seydel et al., 1972). Trimethoprim (TRIM) is used to treat a number of infections including those of the urinary and respiratory tract and belongs to the class of chemotherapeutic agents known as dihydrofolate reductase inhibitors. TRIM acts by targeting an enzyme involved in the tetrahydrafolic acid pathway and so SMX and TRIM have often been used together in therapy since the late 1960s (Burchall, 1973; Seydel et al., 1972). Clarithromycin (CLAR) is used to treat infections such as skin, throat and pneumonia. Azithromycin (AZO) has been used to treat throat, intestinal and sexually transmitted infections. Both of these are macrolide antibiotics which came on the market in the early 1990s and bind to the microbial 50s ribosome sub-unit thereby inhibiting protein synthesis (Piscitelli et al., 1992; Retsema et al., 1987). Tetracycline (TET) is often used in treating skin infections and Lyme disease whilst oxytetracycline (OXY) has been used to treat skin, chest and genital infections. These two antibiotics have been in use since the 1960s and target the microbial 30s ribosomal sub-unit to inhibit protein synthesis (Gale, 1963). Levofloxacin (LEVO) has been used to treat a range of infections including intestinal, pneumonia, urinary tract and prostitis. It belongs to the fluoroquinolone group of antibiotics, which became widely used from 1990, and function by inhibiting the DNA gyrase and topoisomerase IV enzymes (Drlica and Zhao, 1997; Hooper et al., 1987).

By looking at 4 different sewage treatment plants over 4 years, sampling on two occasions each year through taking 24 h composite samples, this study attempted to address the following questions:

How variable are antibiotic effluent concentrations over four years within four different UK sewage treatment plants? To what extent are antibiotic influent and effluent concentrations predictable based on National consumption rates? To what extent does sewage treatment type influence removal performance? How important are flow, removal performance or changes in drug consumption in the variability of antibiotic effluent concentrations?

2. Materials and methods

2.1. Sampling approach at the treatment plants and flow assessment

Four separate STPs in Southern England, UK were examined (Table 1). Two plants were of the activated sludge (AS) type (Ox and Did) and two smaller plants of the trickling filter (TF) type (Ben and Cho). In the UK, AS plants have a hydraulic residence time in the region of 10–14 h and TF plants only 0.5 h. The AS plants handle the most wastewater but TFs are the most numerous (Johnson et al., 2007). A 24 h composite sample was collected by combining hourly samples using an autosampler (Isco Avalanche, Isco 6712, Hach Sigma SD 900 or Bühler Montec Xian 1000). Each treatment plant was sampled, using composite samplers, on two separate occasions during each sampling campaign in June 2012, August 2013, August 2014 and January and August 2015. There have been issues with insufficient or inappropriate sample frequency leading to a number of misinterpretations in understanding the fate of pharmaceuticals in treatment plants (Ort et al., 2010). One example is that in periods of high rainfall both rainwater and elevated groundwater can enter the sewer system, which could have the effect of greatly diluting concentrations of chemicals derived from local households. However, these sampling periods did not occur during periods of high rainfall in the region (Table S1). In addition, this study used carbamazepine as a form of conservative tracer to help avoid misinterpretations of the relative influence of flow versus other losses as described further below.

Unfortunately, daily flow values at the STPs were not available, only the consented flow (Table 1). In the UK, sewage treatment is managed by private companies and there is no requirement to make such information publically available. However, the conservative pharmaceutical carbamazepine (CBZ) makes a very suitable indicator for changes in flow. It is very widely prescribed for epilepsy patients, who take this medication at the same dose throughout the year, and its remarkable persistence has been noted before (Clara et al., 2004; Nakada et al., 2008). Measurements taken in this study showed no decline between the influent and effluent (Tables S4 and S6).

Download English Version:

https://daneshyari.com/en/article/5749520

Download Persian Version:

https://daneshyari.com/article/5749520

Daneshyari.com