Environmental Modelling & Software 62 (2014) 112–127

Contents lists available at ScienceDirect

Environmental Modelling & Software

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

Modelling the occurrence, transport and fate of pharmaceuticals in wastewater systems

Laura J.P. Snip [a], Xavier Flores-Alsina [a, b], Benedek Gy. Plósz [c], Ulf Jeppsson [b], Krist V. Gernaey [a, *]

[a] Department of Chemical and Biochemical Engineering, Technical University of Denmark, Building 229, DK-2800 Kgs. Lyngby, Denmark
[b] Division of Industrial Electrical Engineering and Automation (IEA), Department of Biomedical Engineering (BME), Lund University, Box 118, SE-221 00 Lund, Sweden
[c] Urban Water Engineering, Department of Environmental Engineering, Technical University of Denmark, Building 115, DK-2800 Kgs. Lyngby, Denmark

ARTICLE INFO

Article history:
Received 13 November 2013
Received in revised form
8 July 2014
Accepted 8 August 2014
Available online

Keywords:
Activated sludge modelling
Benchmarking
Influent modelling
Xenobiotic trace pollutants
Micropollutants
Model-based evaluation
Multi-criteria decision making
Organic trace chemicals

ABSTRACT

This paper demonstrates how occurrence, transport and fate of pharmaceuticals at trace levels can be assessed when modelling wastewater treatment systems using two case studies. Firstly, two approaches based on: 1) phenomenology; and, 2) Markov Chains, are developed to describe the dynamics of pharmaceuticals with or without clear administration patterns. Additional simulations also show that sewer conditions might have an important effect on the behaviour of the generated compounds and their metabolites. The results demonstrate that different operating conditions in wastewater treatment plants can have opposite effects on the studied pharmaceuticals, especially when they present co-metabolic or inhibitory behaviour in the presence of biodegradable substrate. Finally, the paper ends with: i) a critical discussion of the presented results; ii) a thorough analysis of the limitations of the proposed approach; and, iii) future pathways to improve the overall modelling of micropollutants.

Software availability

Name of the software: BSM-X
Developers: Laura Snip, Xavier Flores-Alsina, Benedek Gy. Plósz, Ulf Jeppsson and Krist V. Gernaey
Programming language: Matlab 2012b
Availability: The source code for generation of WWTP influent micropollutant files and fate of micropollutants can be obtained for free. Contact Krist V. Gernaey, Department of Chemical and Biochemical Engineering, Technical University of Denmark, Building 229, DK-2800 Kgs. Lyngby, Denmark. E-mail: kvg@kt.dtu.dk

1. Introduction

The term "micropollutants" covers trace organic chemicals such as pharmaceuticals, personal care products and biocides. These compounds are found in low concentrations in some water bodies despite the fact that they are not expected to be present in the environment. In many cases, these substances are found within an organism but they are normally not naturally produced by or expected to be present within that organism. This is the consequence of the increasing number of chemicals (from 50,000 up to 100,000) which are being commercially manufactured by industry, subsequently used in households and finally released to the environment through wastewater (Mackay et al., 2006). The potential adverse effects of micropollutants in aquatic environments have been an object of intensive research during the last years (e.g. Ferrari et al., 2003). In many cases, these pollutants can pose a significant risk on the environment and human health (Ternes et al., 2004a). On aquatic life, such adverse effects can be characterised as spread and upholding antibacterial resistance (Baquero et al., 2008), sex reversal and/or intersexuality (Lange et al., 2009) or reduction of the reproductive behaviour (Coe et al., 2008).

Wastewater treatment plants (WWTPs) are still one of the major disposal pathways for many micropollutants as they are not designed to remove these compounds (Ternes et al., 2004a). However, there are different investigations that demonstrate that a change in operating conditions such as sludge retention time (Clara et al., 2005) can effectively improve the elimination of

* Corresponding author. Tel.: +45 45 25 29 70; fax: +45 45 93 29 06.
E-mail addresses: lasn@kt.dtu.dk (L.J.P. Snip), kvg@kt.dtu.dk (K.V. Gernaey).

http://dx.doi.org/10.1016/j.envsoft.2014.08.007
1364-8152/

micropollutants from the liquid phase by sorption, transformation or biodegradation (Joss et al., 2008). Therefore, comparison of operational/control strategies in WWTPs is a promising tool to test the relative removal effectiveness of these compounds.

The Benchmark Simulation Model (BSM) tools have been developed with the aim of having a platform to objectively compare different control strategies of WWTPs. The main objective of BSM1 was creating a platform for benchmarking carbon and nitrogen removal strategies in activated sludge systems (Copp, 2002). The initial platform evolved into BSM1-P (Gernaey and Jørgensen, 2004), BSM1_LT (Rosen et al., 2004) and finally into BSM2 (Jeppsson et al., 2007; Nopens et al., 2010), which allowed the evaluation of: 1) nitrogen and phosphorus removal strategies; 2) monitoring algorithms; and, 3) plant-wide control strategies. In addition, researchers working within the International Water Association (IWA) Task Group on Benchmarking of Control Strategies for Wastewater Treatment Plants developed other BSM-related spin-off products such as the dynamic influent generator (Gernaey et al., 2011), sensor/actuators/fault models (Rosen et al., 2008) and the different computationally efficient implementations (Rosen et al., 2006) of the Anaerobic Digestion Model No. 1 (ADM1) (Batstone et al., 2002), which have been widely used as stand-alone applications in both industry and academia. Over 400 publications, dealing with BSM platforms (or related material), clearly show the interest for the BSM related models within the scientific community (Gernaey et al., 2014). Nevertheless, these tools have not been developed to include one of the emerging challenges that wastewater engineering is facing nowadays: micropollutants.

Literature offers a wide range of studies with extensive measuring campaigns analysing the occurrence of micropllutants at the inlet of wastewater treatment plants (WWTP) (Göbel et al., 2005; Ratola et al., 2012; Verlicchi et al., 2012). At this moment there are only a few published studies which are trying to describe mathematically how these compounds appear at the inlet of the WWTP. For example, De Keyser et al. (2010), within the framework of the European Research Project Score-PP, developed a stand-alone application that generates time series according to specific (phenomenological/stochastic) release patterns. Influent generators have in general demonstrated to be useful tools for modelling studies since they: i) significantly reduce the cost and workload related to measuring campaigns; ii) fill gaps due to missing data in the influent flow-rate/pollution/temperature profiles; and, iii) create additional disturbance scenarios following the same catchment principles (Gernaey et al., 2011). Regarding micropollutants, modelling the influent dynamics is of the utmost importance as the occurrence at the WWTP inlet shows a high variability (Ort and Gujer, 2005). This variability is also – at least to some extent – present in the effluent of the WWTP, and the peak concentrations can easily exceed the predicted no effect concentration (PNEC) of a compound while the average concentration can be below this limit (Nelson et al., 2011).

Another important factor to consider when modelling micropollutants is the transport within the sewer. Ort et al. (2005) stated that the micropollutant load fluctuations are strongly affected by the sewer system topology and properties. In addition, Plósz et al. (2013) demonstrated the importance of considering in-sewer biotransformation when back-calculating illicit drug consumption rates using end-of-pipe (i.e. WWTP influent) trace organics concentrations and flow data. To our knowledge, there have not been many (modelling) studies assessing the potential of biotransformation or sorption processes within the sewer system. Such processes in the sewer have an important effect on the load of the traditional pollutants arriving at a WWTP (Hvitved-Jacobsen et al., 1998; Ashley et al., 2005). For this reason, we can expect that this effect will also be visible on the micropollutants load. The latter factor might have important implications when back-calculating drug consumption values based on measured WWTP influent micropollutant data.

Finally, it is important to mention fate models as well, when discussing modelling of micropollutants. Most dynamic models describing the fate of micropollutants in a WWTP include pseudo-first order kinetics for biotransformation and sorption/desorption equations (Melcer et al., 1994; Joss et al., 2005; Collado et al., 2012; Lust et al., 2012; Vezzaro et al., 2014). Parameter estimation and calibration/validation of these models are usually performed using batch experiments spiked with the commercially available micropollutant (Ternes et al., 2004b; Joss et al., 2005). These conventional models can be limited in describing the behaviour in full-scale systems due to several reasons. First of all, the presence of substances that facilitate/inhibit removal and/or transformation of these compounds might not be present in these model systems (Grady, 1998). Secondly, the electron dependency (anaerobic, anoxic or aerobic conditions) on the different removal/retransformation rates should be taken into account (Suarez et al., 2010; Plósz et al., 2010a). Thirdly, the retransformation of parent chemicals from e.g. human metabolites and sequestration of parent chemicals in solids can significantly influence the observed removal rate and sorption, respectively (Plósz et al., 2010a, 2012). Fourthly, the total concentration of microorganisms with the metabolic activity to biotransform xenobiotics should be explicitly represented in models (Lindblom et al., 2009).

In view of the different above-mentioned issues, we propose an extension of the current benchmark simulation framework with an additional module dealing with micropollutants. Specifically, the study presented herein will focus on a particular type of micropollutants: pharmaceutical compounds. This additional module is based on the Activated Sludge Model for Xenobiotic trace chemical framework (ASM-X) (Plósz et al., 2010a, 2012), as it includes guidelines on: i) characterisation of the pharmaceutical loading and variability in the loading; ii) assessment of the pharmaceutical fractions in aqueous, solid, retransformable, and sequestered state variables; iii) model identification and calibration using batch experiments; and, iv) model evaluation and validation using full-scale WWTP data. The ASM-X is compatible with the BSM tools as it uses the same modelling approach (see Methods section). Also, the model has been published by Plósz et al. (2010a, 2012, 2013), clearly stating the processes (and equations) as well as the parameter values, making it a transparent model.

The extension of the benchmark simulation framework with pharmaceuticals will result in a set of generic model blocks (influent generation, sewer systems, wastewater treatment plant and controllers) that are upgraded with specific features to realistically describe the behaviour of pharmaceuticals in urban water systems.

The objective of this paper is to demonstrate how occurrence, transport and fate of pharmaceuticals can be assessed when modelling wastewater treatment systems (i.e. catchment, sewer network and wastewater treatment plants). As a result of this combined BSM-ASM-X approach, it is possible to generate pharmaceutical loads based on different information sources (lab experiments, literature data) and different mathematical principles (phenomenological/stochastic). In addition, the proposed approach accounts for some of the physico-chemical processes taking place in the sewer system. Finally, our approach enables a simulation-based investigation of the effect of specific control strategies/operational procedures on the biotransformation, retransformation and sorption rates of the different pharmaceuticals. The paper details the development of the BSM-ASM-X extension and then illustrates the performance of this approach with a number of case studies. These case studies investigate the overall WWTP performance for: i) compounds with different release patterns (three