Science of the Total Environment 601–602 (2017) 874–885

Contents lists available at ScienceDirect

Science of the Total Environment

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

Predicting the fate of micropollutants during wastewater treatment: Calibration and sensitivity analysis

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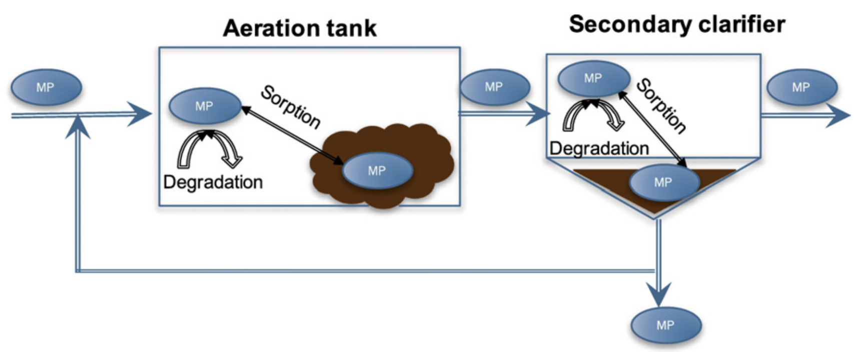
HIGHLIGHTS

- A MP fate model was implemented and calibrated with full-scale measurements.
- Considering biodegradation in the secondary clarifier was important for modelling some MPs.
- The most influential parameters of the model depended on the dominant fate process
- Sensitivity analysis suggests suitable measurement conditions for improved calibration.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 23 December 2016
Received in revised form 11 April 2017
Accepted 8 May 2017
Available online 2 June 2017

Editor: Kevin V. Thomas

Keywords:
Contaminants of emerging concern
Biological treatment
Activated sludge treatment
Dynamic sensitivity
Fate model

ABSTRACT

The presence of micropollutants in the environment and their toxic impacts on the aquatic environment have raised concern about their inefficient removal in wastewater treatment plants. In this study, the fate of micropollutants of four different classes was simulated in a conventional activated sludge plant using a bioreactor micropollutant fate model coupled to a settler model. The latter was based on the Bürger-Diehl model extended for the first time to include micropollutant fate processes. Calibration of model parameters was completed by matching modelling results with full-scale measurements (i.e. including aqueous and particulate phase concentrations of micropollutants) obtained from a 4-day sampling campaign. Modelling results showed that further biodegradation takes place in the sludge blanket of the settler for the highly biodegradable caffeine, underlining the need for a reactive settler model. The adopted Monte Carlo based calibration approach also provided an overview of the model's global sensitivity to the parameters. This analysis showed that for each micropollutant and according to the dominant fate process, a different set of one or more parameters had a significant impact on the model fit, justifying the selection of parameter subsets for model calibration. A dynamic local sensitivity analysis was also performed with the calibrated parameters. This analysis supported the conclusions from the global sensitivity and provided guidance for future sampling campaigns. This study expands the understanding of micropollutant fate models when applied to different micropollutants, in terms of global and local sensitivity to model parameters, as well as the identifiability of the parameters.

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http://dx.doi.org/10.1016/j.scitotenv.2017.05.072

1. Introduction

The presence of pharmaceuticals and personal care products (PPCPs), as well as hormones, in the environment was proven to have adverse effects on aquatic life, raising concern about their insufficient removal from wastewater (Gay et al., 2016; Kidd et al., 2007; Purdom et al., 1994). The two major processes influencing the fate of micropollutants (MPs) during activated sludge treatment are biodegradation and sorption that act at different degrees on MPs with different characteristics (Joss et al., 2006; Radjenović et al., 2009; Ternes et al., 2004). Hence, although most of the emphasis in activated sludge mathematical modelling has been placed on conventional pollutants, some mathematical models were also developed to describe the fate and discharge of MPs in activated sludge systems and proposed as a cost-effective tool for risk assessment (Cowan et al., 1993; Plósz et al., 2012; Urase and Kikuta, 2005; Vezzaro et al., 2014).

Since the fate of MPs is influenced by the fate of conventional pollutants (e.g. solids that affect the sorption and biodegradation of micropollutants), simultaneous modelling of conventional and micropollutants was made possible by extending the well-known Activated Sludge Models (ASMs) developed many years ago (Henze et al., 1987; Henze et al., 1999). The most notable examples of models tackling both conventional pollutants and MPs are the ASM-X model (Plósz et al., 2012; Plósz et al., 2010) and another more recent model based on the ASM2d model (Vezzaro et al., 2014). The majority of MP fate studies in activated sludge focused their efforts on the modelling of the removal in bioreactors and considered the MP fate processes taking place in the secondary clarifier as insignificant (Cloutier et al., 2008; Plósz et al., 2010). However, it is well-known that biological degradation of conventional pollutants can occur in the settler, especially under conditions of long residence times and incomplete denitrification (Siegrist et al., 1995; Koch et al., 1999). Although no specific experimental evidence for the removal of micropollutants in secondary settling tanks has been presented in the literature, MPs are known to co-metabolise with other biodegradable compounds (Clouzot et al., 2013). Hence, the removal of micropollutants in secondary settling tanks should, likely, not be disregarded. So far, only one study by Vezzaro et al. (2014) considered MP fate processes in the clarifier through an extension of the Takács settling model (Takács et al., 1991).

Several constitutive relations to model the different MP fate processes have been proposed in the literature (Plósz et al., 2013; Clouzot et al., 2013). For environmental applications, mostly non-compound specific relations have been used to describe the kinetics of biodegradation, as well as the kinetics and equilibrium of sorption of micropollutants in activated sludge units (Joss et al., 2006; Pomiès et al., 2013). Previous efforts were made to calibrate the model parameters to describe the fate of specific types of MPs (Cloutier et al., 2008; Cowan et al., 1993; Pomiès et al., 2013), with only few studies focusing on PPCPs and hormones (Plósz et al., 2012; Plósz et al., 2010; Urase and Kikuta, 2005). However, a large uncertainty on the calibrated parameters was identified as a major gap in the field of micropollutant modelling in WWTPs, a factor that is aggravated by the lack of knowledge on the sensitivity of the MP fate models to their parameters (Pomiès et al., 2013). This leaves the model users unsure about the degree of confidence in the parameters values reported in the literature.

Measurements of the MP loads sorbed onto sludge for model parameter calibration are limited in the literature to lab scale measurements (Joss et al., 2004; Plósz et al., 2012; Xue et al., 2010), which often cannot be simply extrapolated to the fate of MPs in the complex environment of full-scale WWTP systems. On the other hand, full-scale sampling campaigns are often limited in resources and time preventing very dedicated experiments (Clouzot et al., 2013).

In the present study, a MP fate model based on ASM2d (bioreactor) was further modified and coupled to a reactive settler model extended for the first time from the Bürger-Diehl settler model, which itself incorporates the latest important advancements in the field of secondary settler modelling (Bürger et al., 2012). The scope of the current paper was to obtain reasonable predictions of the removal efficiencies of different types of micropollutants during activated sludge treatment through the calibration of the most influential model parameters for each compound. This was performed using full-scale MP concentration measurements collected from a sampling campaign that was meant to be feasible in terms of efforts and resources supplied by the utility running the plant.

Given the chronic nature of the impacts of the studied CECs, subdaily variations were not deemed of importance for this study, and only the 24-h average load was considered for sampling. Samples were collected under dry weather conditions, since that is when surface water bodies are expected to be the most sensitive to wastewater discharges as a result of the limited dilution. A model that was previously calibrated with respect to conventional pollutants using one-year data was used as a starting point for the current MP calibration. The MPs investigated in the present study included a hormone (i.e. androstenedione), a pharmaceutical (i.e. ibuprofen), an antibacterial agent (i.e. triclosan) and a nervous stimulant (i.e. caffeine). Ibuprofen, triclosan and caffeine were selected based on their high detection frequency in wastewater effluents (Dickenson et al., 2011), while little data is available regarding the levels and fate of androstenedione (Baalbaki et al., 2016; Esperanza et al., 2007). The target MPs were also previously observed to be influenced by sorption and biodegradation fate processes to variable extents in WWTPs (Baalbaki et al., 2016), while volatilization is not considered as significant (Struijs et al., 1991; Virkutyte et al., 2010). For sorption, both the kinetics and the equilibrium equations were considered in the bioreactor and settler models. Sensitivity analysis was performed to explore the impact of the MP fate model parameters on each of the fitted variables, in relation to the input dynamics. Prior to calibrating the model for micropollutant fate, the hydraulic model of the WWTP was identified and the activated sludge unit was calibrated with respect to conventional pollutants, as described in our previous studies (Baalbaki et al., 2016; Baalbaki et al., 2017).

2. Materials and methods

2.1. Full-scale activated sludge

Concentrations of the target MPs were measured in samples collected over four dry days at the full-scale activated sludge unit of the Guelph WWTP (Guelph, Ontario, Canada). The WWTP contains four activated sludge lines, followed by tertiary treatment by rotating biological contactors (RBCs) and sand filtration, as well as disinfection by chlorine. It serves a population of 135,000 inhabitants and receives an average flow of 50,750 m^3/day. The final de-chlorinated effluent is discharged into a nearby river. The first of the four lines of activated sludge (line 1) was selected for this study. This line contains two aeration tanks in parallel, and the output of both tanks is combined and sent to a single secondary clarifier. Ferric chloride is added for phosphorus removal to the input to the primary clarifier (i.e. 1 L/min) and at a lower dosage to the return sludge of line 1 (i.e. 0.55 L/min). The main characteristics of line 1, including average hydraulic and solids retention time (HRT and SRT, respectively), as well as the average mixed liquor suspended solids concentration (MLSS), average mixed liquor volatile suspended solids concentration (MLVSS) and average temperature measurements over the sampling period are summarized in Table 1.

2.2. Sampling

Sampling was performed over four days in the period of July 21–25, 2014 in dry conditions. The number of sampling days was selected based on a hydraulic model for this WWTP (Baalbaki et al., 2016). As shown in Fig. 1, samples were collected from the effluent of the primary clarifier or primary effluent (PE), the effluent of the aeration tanks (AE), the effluent of the secondary clarifier (SE), as well as from the secondary