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Assessment of the fate of organic micropollutants in novel wastewater treatment plant configurations through an empirical mechanistic model

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Abstract

Novel wastewater treatment plants (WWTPs) are expected to be less energetically demanding than conventional ones. However, scarce information is available about the fate of organic micropollutants (OMPs) in these novel configurations. Therefore, the objective of this work is to assess the fate of OMPs in three novel WWTP configurations by using a plant-wide simulation that integrates multiple units. The difference among the three configurations is the organic carbon preconcentration technology: chemically enhanced primary treatment (CEPT), high-rate activated sludge (HRAS) combined or not with a rotating belt filter (RBF); followed by a partial-nitritation (PN-AMX) unit. The simulation results show that the three selected novel configurations lead mainly to comparable OMPs removal efficiencies from wastewater, which were similar or lower, depending on the OMP, than those obtained in conventional WWTPs. However, the presence of hydrophobic OMPs in the digested sludge noticeably differs among the three configurations. Whereas the configuration based on sole HRAS to recover organic carbon leads to a lower presence of OMPs in digested sludge than the conventional WWTP, in the other two novel configurations this presence is noticeable higher. In conclusion, novel WWTP configurations do not improve the OMPs elimination from wastewater achieved in conventional ones, but the HRAS-based WWTP configuration leads to the lowest presence in digested sludge so it becomes the most efficient alternative.

Keywords: biotransformation, chemically enhanced primary treatment, high-rate activated sludge, plant-wide simulation, rotating belt filters, thermal hydrolysis.

1. Introduction

Conventional wastewater treatment plants (WWTPs) are expected to be replaced by a new generation, which offers up to 60% reduction in aeration requirements and consequently in energy consumption. This fact allows the WWTPs to approach the energy autarky or even become net electrical producers (Gikas, 2017; Gu et al., 2017; Wan et al., 2016). In novel WWTPs, chemical oxygen demand (COD) is recovered in a first stage followed by a partial nitritation-anammox (PN-AMX) unit. Several pre-concentration alternatives can be applied, such as rotating belt filters (RBF), chemically enhanced primary treatment (CEPT), high-rate activated sludge (HRAS) or combinations thereof (Lotti et al., 2014). The COD recovered as sludge is pretreated through different technologies, such as thermal hydrolysis (TH), to increase biogas yield and reduce sludge production after anaerobic digestion (AD) (Sapkaite et al., 2017).

A great effort has been made over the last two decades to determine the occurrence of OMPs in wastewater and their fate in the different units of conventional WWTPs, such as primary clarifiers (Behera et al., 2011; Carballa et al., 2004), activated sludge reactors (Alvarino et al., 2014; Radjenović et al., 2009; Santos et al., 2009; Suarez et al., 2010) or anaerobic digesters (Gonzalez-Gil et al., 2016; Narumiya et al., 2013; Yang et al., 2016). However, these novel technologies are yet at their early stages of industrial implementation, although preliminary works studying the fate of OMPs in RBF systems (Taboada-Santos et al., 2019b), HRAS reactors and CEPT (Taboada-Santos et al., 2020), PN-AMX reactors (Alvarino et al., 2015; Kassotaki et al., 2018; Laureni et al., 2016) or sludge TH (Reyes-Contreras et al., 2018; Taboada-Santos et al., 2019a; Zhang and Li, 2018) can already be found in the literature. However, these units are commonly studied individually, so it is essential to integrate multiple units to holistically assess the fate of OMPs in novel WWTPs. Plant-wide simulation can be an appropriate approach since it has been successfully applied in wastewater treatment, mainly focused on energetic and/or economic aspects (Behera et al., 2018; Flores-Alsina et al., 2014, 2011; Mbamba et al., 2019). Few full-scale modelling studies are also available on OMPs removal in conventional WWTPs (Lautz et al., 2017; Polesel et al., 2016; Pomiès et al., 2013; Snip et al., 2014; Struijs et al., 2016; Xue et al., 2010). However, to the best of our knowledge, there are not works available on the fate of in novel configurations of WWTPs. The goal of this work is to evaluate the fate of OMPs in novel WWTP configurations by using an empirical mechanistical model. The results obtained were compared with the fate of OMPs in conventional WWTPs.

2. Materials and methods

2.1. Novel WWTP configurations

Three novel WWTP configurations based on HRAS (Figure 1A), a combination of RBF and HRAS (Figure 1B) and CEPT (Figure 1C) for COD capture, followed by a mainstream PN-AMX were considered. The sludge line was common for the three configurations: a sludge thickener, a TH unit, an anaerobic digester and a dewatering unit.

A fourth configuration, representing a conventional WWTP based on conventional primary treatment (CPT) + conventional activated sludge (CAS) in the water line and thickener + anaerobic digester + dewatering in the sludge line was also included (Figure 1D).

2.2. Plant-wide modelling

The WWTP size considered for the plant-wide analysis was 100,000 inhabitants equivalents with an average flowrate of 20,800 m^3/d (Gernaey et al., 2011).

2.2.1. CPT, CEPT and RBF systems

The CPT was modelled based on the gravity settling principle by which heavier solids settle down faster. The performance of primary clarifiers can be enhanced by the addition of chemicals or polymers, known as CEPT, which boost not only the particulate matter but also the soluble matter removal. The CEPT unit was modelled as an ideal separator where, by the addition of 125-150 mg/L of ferric chloride, particulate COD matter removal was set to 99% and the soluble COD fraction was set to 50-60% (Taboada-Santos et al., 2019b).The RBF unit works based on cake filtration and sieving. It was modelled as described elsewhere (Behera et al., 2018; Boiocchi et al., 2019).

2.2.2. HRAS and CAS reactors

The HRAS which works on bio-sorption principle was modelled as a continuous stirred-tank reactor (CSTR) followed by a settler (Smitshuijzen et al., 2016). The hydraulic retention time (HRT) and solid retention time (SRT) were set to 30 min and 0.3 d, respectively. The dissolved oxygen (DO) concentration inside the reactor was set to 0.2 mg/L to avoid unnecessary oxidation of biodegradable COD.

Likewise, the CAS unit was modelled as a Modified Ludzack-Ettinger (MLE) system with two anoxic tanks (for pre-denitrification) and three aerobic tanks (for nitrification) (Gernaey et al., 2014), followed by a settler. The activated sludge model ASMG1 (Guo and Vanrolleghem, 2014) was used to model both tanks. The DO in the aerobic tanks was maintained at 1 mg/L and a constant addition of external carbon (800 kg/d) to anoxic tanks was assumed for complete denitrification. A HRT of 21 hours and a SRT of 14 days were maintained in CAS system to ensure efficient nitrification.

The settler of both CAS and HRAS was modelled as a 10 layers non-reactive settling tank using the exponential settling velocity function proposed by Takács et al. (1991).

2.2.3. PN-AMX reactor

The PN-AMX unit was considered as integrated fixed film activated sludge system (IFAS), a promising technology for mainstream nitrogen removal application

(Malovanyy et al., 2015). The IFAS system is modelled using a multiscale approach where the carrier geometry was assumed to be a flat sheet. The biofilm growth was simplified to one dimensional, a commonly used approach in other studies (Eberl et al., 2006; Lindblom et al., 2016; Vangsgaard et al., 2013). A relatively low DO (0.1 mg/L) compared to CAS system was maintained to suppress the nitrite oxidizing bacteria growth (Cao et al., 2017; Malovanyy et al., 2015).

2.2.4. TH and AD units

The TH unit was modelled by converting inert and slowly biodegradable particulate COD to soluble biodegradable COD (Bougrier et al., 2008) in the same percentage as anaerobic biodegradability increased after TH, according to Taboada-Santos et al. (2019c).

The anaerobic digester was modelled using ADM1 (Batstone et al., 2002), assuming a SRT of 19 days in all configurations (Gernaey et al., 2014).

2.2.5. Thickening and dewatering units

The thickening and the dewatering units were modelled using a constant thickening and dewatering factor (Gernaey et al., 2014).

2.3. Incorporation of OMPs to the plant-wide model

2.3.1. Raw wastewater

Most of the authors in the literature disregard the solid phase when they determine the occurrence of OMPs in the influents of WWTPs. However, in this work, both liquid and solid phases were considered in order to perform a more sensitive analysis. Total OMPs concentration in a stream (C_t, mg/m^3) is normally expressed as the sum of its soluble concentration (C_w, mg/m^3) and its sorbed concentration (C_s, mg/m^3) (Eq. 1).

$$C_t = C_w + C_s \tag{1}$$

A common approach to determine the fraction of OMPs sorbed onto suspended solids is the use of the solid–water distribution coefficient (K_D, m^3/kg TSS), defined as the ratio between the concentrations in the solid and liquid phases at equilibrium conditions (Eq. 2).

$$K_D = \frac{C_s}{C_w \cdot TSS} \tag{2}$$

Where (TSS, kg/m^3) is the total suspended solids concentration in that stream.

Combining Eq.1 and Eq. 2, C_t, can be obtained by Eq. 3.

$$C_t = C_w + TSS \cdot K_D \cdot C_w \tag{3}$$

2.3.2. CPT, RBF and CEPT units

The fate of OMPs in the physico-chemical separation units was modelled assuming that no biodegradation occurred, so the removal of OMPs in these units is attributed to TSS separation (Carballa et al., 2004). As sorption depends on several factors, such as the physico-chemical properties of TSS, the chemicals involved or the ambient conditions (pH, ion strength, temperature, etc) (Carballa et al., 2008), different K_D values in CPT, RBF an CEPT sludges were considered, and soluble and particulate concentrations were calculated by Eq. 1-3.

2.3.3. CAS, HRAS and PN-AMX units

Considering pseudo steady-state conditions and assuming a CSTR and negligible volatilisation as previously stated (Alvarino et al., 2014), the following mass balance can be established (Eq. 4) in any biological reactor (CAS, HRAS and PN-AMX):

$$F_{biod} = F_{inf} - F_{eff} - F_s \tag{4}$$

where F_{inf}, F_{eff}, and F_s represent the mass flows (in mg/d) corresponding to the influent, effluent and the purged sludge. F_{inf} F_{eff} and F_s can be expressed as the product of the flowrate ($F_{R,inf}$, $F_{R,eff}$, $F_{R,s}$, m^3/d) by the total OMP concentration in that stream ($C_{t,inf}$, $C_{t,eff}$, $C_{t,s}$, mg/m^3), respectively (Eq. 5).

$$F_{biod} = F_{R,inf} \cdot C_{t,inf} - F_{R,eff} \cdot C_{t,eff} - F_{R,s} \cdot C_{t,s} \quad (5)$$

Assuming a pseudo-first kinetic biotransformation, the flux of biotransformed OMP (F_{biod}, mg/d) can be expressed as shown in Eq. 6.

$$F_{biod} = k_{biol} \cdot VSS \cdot C_{t,eff} \cdot V \quad (6)$$

where k_{biol} ($m^3/kg_{VSS} \cdot d$) represents the pseudo-first order kinetic constant, VSS is the biomass concentration in the reactor (kg VSS/m^3) and V is the reactor volume (m^3). Assuming that soluble OMP concentration in the effluent ($C_{w,eff}$, mg/m^3) and in sludge ($C_{w,sl}$, mg/m^3) is exactly the same and that the liquid and solid phase of each stream are in equilibrium, the $C_{w,eff}$ can be calculated by Eq. 7.

$$C_{w,eff} = \frac{C_{t,inf} \cdot F_{R,inf}}{k_{biol} \cdot VSS \cdot V \cdot \left(1 + TSS_{eff} \cdot K_D\right) + F_{R,eff} \cdot \left(1 + TSS_{eff} \cdot K_D\right) + F_{R,s} \cdot (1 + TSS_s \cdot K_D)} \quad (7)$$

where K_D (m^3/kg TSS) is the OMP solid-liquid equilibrium constant in the biological sludge, TSS_{eff} and TSS_s (kg/m^3) the TSS concentration in the effluent and in waste sludge, respectively. From $C_{w,eff}$, sorbed concentration in the effluent and in the sludge can be calculated by Eq. 2.

2.3.4. Sludge thickener

The fate of OMPs during sludge thickening was modelled assuming that there is no variation in soluble neither specific sorbed OMPs concentration (µg/g of TSS). Therefore, the total OMP concentration in thickened sludge ($C_{t,thick}$, mg/m^3) was calculated by Eq. 1.

2.3.5. TH unit

It is well known that TH causes a partial solubilisation of particulate solids and organic matter; however, the information in the literature assessing the fate of OMPs in TH plants is quite scarce. A recent study carried out by Taboada-Santos et al. (2019a) found that after TH the sorbed OMPs concentration in sludge ($C_{s,pt}$, mg/m^3) was reduced with respect to that in the influent ($C_{s,fresh}$, mg/m^3) in the same percentage as TSS were solubilised, and can be calculated by Eq. 8.

$$C_{s,pt} = C_{s,fresh} \cdot \frac{TSS_{pt}}{TSS_{fresh}} \quad (8)$$

Being TSS_{fresh} (kg TSS/m^3) the TSS of sludge before TH and TSS_{pt} (kg TSS/m^3) the TSS of pretreated sludge.

They also found that the soluble (and solubilised) concentrations of some OMPs decreased during TH. Therefore, OMPs soluble concentration in pretreated sludge can be calculated by Eq. 9.

$$C_{w,pt} = \left(C_{w,fresh} + C_{s,fresh} \cdot \frac{TSS_{fresh} - TSS_{pt}}{TSS_{fresh}}\right) \cdot (1 - R) \quad (9)$$

Being R (0-1) the removal of soluble and solubilised OMPs achieved during TH. Total OMPs concentration in pretreated sludge ($C_{t,pt}$, mg/m^3) is subsequently calculated as the sum of its soluble and particulate concentration (Eq. 10).

$$C_{t,pt} = C_{s,fresh} \cdot \frac{TSS_{pt}}{TSS_{fresh}} + \left(C_{w,fresh} + C_{s,fresh} \cdot \frac{TSS_{fresh} - TSS_{pt}}{TSS_{fresh}}\right) \cdot (1 - R) \quad (10)$$

2.3.6. AD unit

Contrary to the mainstream biological units, the fate of OMPs during sludge AD was not modelled as a pseudo-first kinetics, since in a recent study Gonzalez-Gil et al. (2018) found that OMPs biotransformation during AD is likely limited by thermodynamic rather than kinetic constraints, and using pseudo-first order kinetics could lead to an overestimation of the biotransformation capacity. Therefore, for modelling this unit, a fixed OMPs biodegradability (B_t) was considered, and the total OMPs concentrations in digested sludge ($C_{t,dig}$, mg/m^3) was calculated by Eq. 11.

$$C_{t,dig} = C_{t,feed} \cdot (1 - \frac{B_t}{100}) \quad (11)$$

Being $C_{t,feed}$ the total OMPs concentration (mg/m^3) in the anaerobic digester feeding.

The soluble and sorbed OMPs concentration in digested sludge can be calculated by Eq. 1-3.

2.3.7. Digested sludge dewatering

The fate of OMPs in the digested sludge dewatering unit was modelled as previously explained in section 2.3.3. for the sludge thickener.

2.4. Selection of OMPs and data input for the model

Seventeen compounds commonly used in daily life were considered in this study: three musk fragrances, galaxolide (HHCB), tonalide (AHTN) and celestolide (ADBI); three anti-inflammatories, ibuprofen (IBP), naproxen (NPX) and diclofenac (DCF); four anti-biotics, sulfamethoxazole (SMX), trimethoprim (TMP), erythromycin (ERY) and roxithromycin (ROX); three neurodrugs, fluoxetine (FLX), carbamazepine (CBZ), diazepam (DZP); one endocrine disrupting compound, triclosan (TCS); and three hormones, estrone (E1), 17β-estradiol (E2) and 17α-ethinylestradiol (EE2).

The occurrence of OMPs in urban wastewater is quite wide, and OMPs concentrations in the influent were selected in the range of the values reported by Luo et al. (2014) and Verlicchi et al. (2012); 1 ppb for estrogens and 10 ppb for the rest of compounds. As previously indicated, both soluble and sorbed fractions of OMPs in the influent were considered. Figure S1 shows the relative presence in the liquid and solid phase of the 17 selected OMPs for this study for an influent with 380 mg/L of TSS.

2.4.1. Solid-liquid distribution coefficient (K_D) of OMPs in the different sludges

The technology selected to recover organic matter strongly affects the nature of the sludge produced (i.e. RBFs mainly captures cellulose, CEPT captures not only particulate matter but also soluble one, etc.) and might lead to different solid-liquid equilibrium coefficients of OMPs. Therefore, for each sludge, different coefficients were considered to take the sludge characteristics into account. Table 1 shows the K_D

values for the different sludges considered in this work found in the literature and the most representative value, which was the one used to carry out this work.

2.4.2. Pseudo-first order biotransformation constants (k_{biol}) of OMPs in the different main-stream biological reactors

Table 2 displays the k_{biol} values found in the literature for the different biological units considered in this work, and the most representative value, which was the one used to carry out this work.

The number of studies in CAS based on the nitrification-denitrification process is huge; however, for HRAS and PN-AMX reactors, the information is still scarce (Table 2) and only one work for each technology was found in the literature, so the modelling assumptions included in this paper should be further supported with additional experimental work around the technologies considered. Moreover, for the PN-AMX technology the only study found studied the fate of OMPs in a reactor treating the sludge supernatant rather than in mainstream conditions (Alvarino et al., 2015), . However, a recent study from Laureni et al. (2016) reported, for some OMPs, very comparable removal efficiencies in a CAS and a mainstream PN-AMX reactor, and considering that the k_{biol} values reported by Alvarino et al. (2015) were in the same range of those found for CAS, they were taken as representative for mainstream PN-AMX unit.

2.4.3. OMPs removal in sludge TH and AD

The range of removal efficiencies of OMPs during AD in the literature is quite wide, and sometimes controversial. Table 3 summarises the results found in the literature for the selected OMPs and the representative values considered in this work. The removal efficiency of the soluble and solubilised fraction of OMPs during sludge TH was taken from Taboada-Santos et al. (2019a).

2.5. Sensitivity analysis

The prediction of the fate of the OMP in the WWTP depends critically on the values of the parameters governing the kinetics of OMP biotransformation and the phase equilibria. We performed a global uncertainty and sensitivity analysis of the input parameter space, the values of the kinetic and equilibrium parameters as well as the biotransformation efficiency during AD with two goals: i) estimating the uncertainty associated to the simulations of the fate of OMPs and ii) identifying the most sensitive parameters for the simulations. From these two results it is possible to decide whether the uncertainty of the predictions is acceptable for a future design and, if this uncertainty was to be reduced, the experimental campaign should focus first on the parameters identified as most sensitive for the simulation outcome (Sin et al., 2009).

All the parameters related to OMP fate are considered to be uncorrelated and, furthermore, that the fate of a given OMP does not influence the rest. Their expected value and uncertainty were approximated as the mean and standard deviation of the values in Tables 1-3 of the literature review. The parameter space was sampled using the Latin Hypercube Sampling methods to ensure a maximal coverage of the parameter space (Helton and Davis, 2003). Following a Monte Carlo procedure, each of the scenarios was simulated 400 times and the OMP concentration in the effluent and digested sludge were recorded. These model outputs were the basis for the subsequent sensitivity analysis. It was considered that the behaviour of the plants with respect to nutrients, solids and COD was perfectly known and therefore the rest of parameters were not included in the uncertainty analysis.

A global sensitivity analysis was carried out to determine what parameters have a higher influence on the effluent and sludge OMP content. The method of standardised regression coefficients (SRC) was chosen, consisting on fitting a first order linear multivariable model between the predictions and the parameter values (θ_i) by a least squares method (Saltelli et al., 2008):

$$y_k = b_{k,0} + \sum_i b_{k,i} \cdot \theta_i \tag{12}$$

where y_k are the content of OMP k in a given stream, $b_{k,0}$ and $b_{k,i}$ are the linear regression coefficients and θ_i the parameters, with index k varying from 1 to the number of OMP and index i from 1 to the number of parameters. To assume the model linear, the squared coefficient of correlation (R^2) between the Monte Carlo simulation output (Y) and the values produced with the regression model with the estimated SRC (Eq. 9) regressed linear output should be above 0.7 (Vangsgaard et al., 2012), which was confirmed for all the cases analysed. After standardisation of the outputs and parameters, the absolute magnitude of the regression coefficients indicates the sensitivity of the outputs to a given parameter and, therefore, can be used to rank the parameters with a higher influence on the predictions. Only those parameters with an expected influence larger than 5% were retained for further analysis.

3. Results and discussion

3.1. Aeration demand, methane production and effluent quality of novel WWTP configurations

As expected, novel configurations lead to considerably lower aeration demand than the conventional configuration (Table 4). This is primarily because of the implementation of the mainstream PN-AMX which greatly reduces the energy consumption compared to the CAS reactor, supporting other studies findings (Cao et al., 2017; Malovanyy et al., 2015). Moreover, lower TN concentrations in the effluent are achieved in the novel configurations, since nitrate removal is only partial in the conventional configuration due to insufficient COD.

Novel configurations also achieve considerable higher methane production than the conventional alternative, not only due to the higher COD recovery from wastewater but also due to the increase of methane productivity after sludge pretreatment. Regarding sludge production, it also results higher in novel configurations, as previously reported by Taboada-Santos et al. (2019c).

3.2. Removal efficiency of OMPs from the water line in novel WWTP configurations

Figure 2 shows the removal efficiency of the selected OMPs from wastewater (attributed to biotransformation and sorption into sludge) in the three novel WWTPs configurations and also in the conventional one. For compounds, such as AHTN, HHCB, ADBI, TCS, E1+E2, IBP and NPX, high removal efficiencies (>70%) were found in both novel and conventional configurations. Other OMPs, such as TMP, DZP, CBZ and DCF also presented similar removal efficiencies in novel and conventional WWTPs configurations, but with lower values (< 40%). Finally, E2, FLX, ROX, SMX and ERY displayed much lower removal efficiencies in novel WWTPs (between 13% and 61%) than in the conventional ones (between 84% and 95%). The parameters with an expected influence larger than 5% on the OMP removal efficiency were retained for the analysis and are shown in Table S1 in the Supporting Information. In novel WWTPs configurations it was found that, for hydrophilic OMPs, k_{biol} in both the HRAS and PN-AMX reactors are the only parameters that influence the OMP removal from wastewater, whereas the K_D value in the HRAS sludge is also relevant for hydrophobic compounds in the HRAS- and RBF+HRAS-based configurations. Contrary, in the conventional configuration, the k_{biol} value in CAS reactors is the only parameter that plays a significant role for most of the OMPs regardless their hydrophobicity.

The lower removal efficiencies obtained in novel configurations can be attributed to two reasons. First, the low HRT applied in HRAS reactors to minimize COD mineralization, since for most of them medium or high k_{biol} values were obtained by Taboada-Santos et al. (2019d), indicating that their biotransformation is limited by the low HRT applied. Furthermore, according to Jimenez et al. (2005), particulate and colloidal COD is removed from wastewater by biological flocculation and subsequent settling, whereas the soluble fraction is eliminated by intracellular storage, biosynthesis or biological oxidation. Therefore, less COD is metabolized in this unit than in CAS reactors, producing a reduction of co-metabolism activity, which is thought to be the main mechanism for OMPs biotransformation (Gauthier et al., 2010; Kassotaki et al., 2016). Second, whereas in CAS units it is desired that 100% of the ammonia is converted to nitrite and afterwards to nitrate, in PN-AMX units only the 50% of ammonia is oxidized to nitrite. Considering again a co-metabolic approach, this lower ammonia oxidation could result on a lower biotransformation efficiency of OMPs. Moreover, the lack of nitrite oxidizing microorganisms in PN-AMX reactors might limit OMPs biotransformation. Even though some works in the literature indicate that the removal of OMPs in CAS is linked to nitrifying activities (Fernandez-Fontaina et al., 2016), other works suggest that there is a potential overestimation of the contribution of ammonia oxidizers to OMP biotransformation to the detriment of nitrite oxidizers (Men et al., 2017).

3.3. Fate of OMPs in novel WWTP configurations

According to the fate of selected OMPs in novel WWTP configurations, they were classified into four groups.

Group I: Hydrophobic OMPs (log K_D ≥3.5)

Hydrophobic OMPs, such as AHTN, HHCB, ADBI and TCS, are well eliminated from wastewater, attaining removal efficiencies between 73% and 88% (Figure 2). Although the removal efficiencies were quite comparable in the three configurations, important differences were found regarding their fate (TCS, was selected as representative of this group of OMPs in Table 5). The HRAS-based configuration is the alternative that leads to the lowest flux in the final effluent and digested sludge (Table 5). This is due to the high biotransformation efficiency of TCS (up to 43%) achieved in the HRAS reactor (Figure S2), even with the very low HRT (30 min) applied, attributed to its very high k_{biol} value under heterotrophic conditions (Table 2). In contrast, the PN-AMX unit did barely contribute to biotransform TCS (<4%, Figure S2). Additionally, 41% of TCS is removed from wastewater sorbed into sludge (Figure S2), attributed to its high hydrophobic behaviour, but its presence in the digested sludge (Table 5) is reduced due to its medium biotransformation efficiency during AD.

The partial TSS removal achieved in the RBF causes that approximately 33% of TCS in the influent is diverted to the sludge line before reaching the biological units (Figure S3). As a consequence, its biotransformation efficiency decreases to 25%, although not affecting the mass flux in the final effluent (Table 5), whereas removal by sorption into sludge increases up to 60% (Figure S3). Therefore, a slightly higher mass flux in digested sludge is obtained in this configuration (Table 5).

A slightly higher effluent mass flux is obtained in the CEPT-based configuration (Table 5) attributed to the lack of a HRAS reactor The high TSS elimination achieved in the CEPT unit produces a removal efficiency of almost 80% due to sorption (Figure S4). Subsequently, its mass flux in digested sludge is the highest one (Table 5).

Group II: Hydrophilic OMPs (log $K_D \leq 3.2$) with $k_{biol} \geq 10$ L/g $_{VSS}$·d in the HRAS reactor and/or ≥5 L/g $_{VSS}$·d in the PN-AMX reactors.

This group includes those hydrophilic OMPs that present high k_{biol} values in the HRAS and/or in the PN-AMX reactors, such as E1, E2, IBP and NPX. These compounds are well biotransformed in both biological units reaching removal efficiencies from wastewater above 95% in the three configurations. IBP was selected as representative of this group in Table 5.

The mass fluxes in the final effluent (Table 5) are very comparable in the three configurations, which is explained by the high biotransformation efficiencies in both biological systems, 69-79% in the HRAS unit and 93-94% in the PN-AMX one. In the HRAS- and RBF+HRAS-based configurations, approximately 70-80% of IBP is biotransformed under heterotrophic conditions, noticeably reducing the mass flux that reaches the PN-AMX reactor (Figure S2 and S3), in which just 20-30% of the OMP in the influent is biotransformed (Figure S2 and S3).

Contrary, in the CEPT-based configuration the PN-AMX reactor biotransforms 94% of the IBP in the influent (Figure S4), fact might be important since different transformation products (TP) are formed due to the different mechanisms involved (heterotrophic or ammonium oxidizer biomass) in the different configurations, and these TP might present different k_{biol} values and/or toxicity (Collado et al., 2012).

The presence of these OMPs in the digested sludge is very low in all configurations (Table 5) since sorption into sludge hardly contributes to their removal from wastewater (Figures S1-S3).

Group III: Hydrophilic OMPs with k_{biol}<10 L/g $_{VSS}$·d in the HRAS reactor and (1≤ k_{biol} <5 L/g $_{VSS}$·d) in the PN-AMX reactor.

This group includes those hydrophilic OMPs which are partially or not removed in the HRAS reactor but show a medium-high removal efficiency in the PN-AMX one, such as EE2.

Similarly to the previous group, the presence of these OMPs in the digested sludge is very low in all configurations (Table 5) since sorption into sludge barely contributes to their removal from wastewater. Regarding the water line, no major differences in their removal were found among the different novel WWTP configurations (Table 5), achieving removal efficiencies of approximately 50-60%, mainly due to the PN-AMX reactor, since the biotransformation efficiency in the HRAS reactors results below 15% (Figure S2 and S3).

Group IV: Hydrophilic OMPs with k_{biol} <10 L/g_{VSS}·d in the HRAS and k_{biol} <1 L/g_{VSS}·d in the PN-AMX reactors.

This group contains those hydrophilic OMPs that are not removed neither in the HRAS nor in the PN-AMX reactors (Figure S2-S4) such as ROX, SMX, ERY, TMP DZP, CBZ and DCF, so they show a recalcitrant behaviour. CBZ was selected as the compound representative of this group. Due to their hydrophilic behaviour, sorption does not contribute to their removal (Figure S2-S4), so their presence in digested sludge is very low in all configurations (Table 5). Medium-low biotransformation efficiencies from wastewater (between 0 and 40%) are obtained, so they achieve a noticeable presence in the WWTPs effluents (Table 5). It must be highlighted that for part of the OMPs of this group including ROX, SMX or DZP, their biotransformation efficiency in the HRAS- and RBF+HRAS-based WWTPs could be enhanced to comparable values to those obtained in the CAS in the conventional configuration (Figure S5) by increasing the HRT, since they present medium k_{biol} values in the HRAS reactor, demonstrating that their biotransformation is limited by the low HRT. However, increasing the HRT would lead to a lower methane production and therefore energy recovery due to a higher COD oxidation, as reported by Jimenez et al. (2015).

It must be highlighted that the k_{biol} values considered for the HRAS reactor were obtained with a DO concentration of approximately 3 mg O_2/L (Taboada-Santos et al., 2020), whereas this model suggests to decrease it in order to minimize COD oxidation. This variation could lead to lower k_{biol} values and therefore different biotransformation efficiencies, proving that more experimental works should be carried out in order to validate the assumptions made in this work.

3.4. Fate of OMPs in the sludge line of WWTPs

Not only the removal from wastewater but also the presence of OMPs in sludge is an important issue, particularly when sludge is used as fertilizer in agriculture. Important differences were found among the novel scenarios, being the CEPT-based configuration the alternative reaching the highest OMP load in the sludge line (Figure S4) and the HRAS-based configuration the lowest one (Figures S2). Again, the parameters with an expected influence larger than 5% on the OMP removal efficiency were retained for the analysis and are shown in Table S2 in the Supporting Information. The K_D coefficient of OMPs in anaerobic sludge influences the presence of most of them in digested sludge in both novel and conventional configurations. Besides, other parameters such as the k_{biol} and K_D values in the different mainstream biological units can be relevant in some cases, since they might significantly impact the presence of OMPs in the sludge line and therefore in digested sludge.

Besides increasing biogas production in AD, TH contributes to a partial removal of OMPs, linked to TSS solubilisation (Taboada-Santos et al., 2019a). This is especially relevant for hydrophobic compounds (Group I), attaining mass fluxes reduction of 26% in the HRAS-based configuration (Figure S1), 32% in the RBF+HRAS-based configuration (Figure S2) and 18% in the CEPT- based alternative (Figure S3). However, the low-medium anaerobic biodegradability reported in the literature for this group of compounds (Table 3) causes that AD only contributes to a partial removal from sludge. Consequently, most OMPs are present in the digested sludge (Figure 3), but this presence of course depends on the characteristics of each specific compound, mainly hydrophobicity and anaerobic biotransformability. For hydrophilic compounds, less than 6% of the influent mass flow is present in digested sludge, whereas for hydrophobic compounds, this number can increase up to 40% (Table 5).

Therefore, this paper gives a first insight about the fate of OMPs in novel schemes for wastewater treatment, but more experimental works should be carried out to obtain more data for the inputs of the model that allow to achieve more robust results.

4. Conclusions

In general, the technology selected for organic matter recovery in novel WWTP configurations does not influence the removal efficiency of OMPs from wastewater, which was found comparable for most of them. Moreover, these novel configurations achieve, depending on the OMP, comparable or lower removal efficiency than a conventional WWTP configuration. However, the organic matter recovery technology determines the presence of hydrophobic OMPs in the sludge line, and subsequently, in the digested sludge. Whereas the HRAS-based WWTP achieves comparable or even lower OMPs presence in digested sludge than the conventional configuration, in the HRAS+RBF and mainly CEPT-based alternatives, their presence is expected to be considerably higher. Therefore, the HRAS-based WWTP configuration is the preferable option in terms of OMPs elimination.

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References

Abegglen, C., Joss, A., Mcardell, C.S., Fink, G., Schlu, M.P., Ternes, T.A., Siegrist, H., 2009. The fate of selected micropollutants in a single-house MBR. Water Res. 43, 2036–2046. https://doi.org/10.1016/j.watres.2009.02.005

Alvarino, T., Suarez, S., Katsou, E., Vazquez-Padin, J., Lema, J.M., Omil, F., 2015. Removal of PPCPs from the sludge supernatant in a one stage nitritation/anammox process. Water Res. 68, 701–709. https://doi.org/10.1016/j.watres.2014.10.055

Alvarino, T., Suarez, S., Lema, J.M., Omil, F., 2014. Understanding the removal mechanisms of PPCPs and the influence of main technological parameters in anaerobic UASB and aerobic CAS reactors. J. Hazard. Mater. 278, 506–513. https://doi.org/10.1016/j.jhazmat.2014.06.031

Batstone, D.J., Keller, J., Angelidaki, I., Kalyuzhnyi, S. V, Pavlostathis, S.G., Rozzi, A., Sanders, W.T.M., Siegrist, H., Vavilin, V.A., 2002. The IWA Anaerobic Digestion Model No 1 (ADM1). Water Sci. Technol. 45, 65–73.

Behera, C.R., Santoro, D., Gernaey, K. V., Sin, G., 2018. Organic carbon recovery modeling for a rotating belt filter and its impact assessment on a plant-wide scale. Chem. Eng. J. 334, 1965–1976. https://doi.org/10.1016/j.cej.2017.11.091

Behera, S.K., Kim, H.W., Oh, J.E., Park, H.S., 2011. Occurrence and removal of antibiotics, hormones and several other pharmaceuticals in wastewater treatment plants of the largest industrial city of Korea. Sci. Total Environ. 409, 4351–4360. https://doi.org/10.1016/j.scitotenv.2011.07.015

Bergersen, O., Hanssen, K.Ø., Vasskog, T., 2012. Anaerobic treatment of sewage sludge containing selective serotonin reuptake inhibitors. Bioresour. Technol. 117, 325–332. https://doi.org/10.1016/j.biortech.2012.04.086

Boiocchi, R., Behera, C.R., Sherrat, A., DeGroot, C.T., Gernaey, K. V., Sin, G., Santoro, D., 2019. Dynamic model validation and plant-wide assessment of rotating belt filtration as primary treatment of domestic wastewaters. Submitt. to Chem. Eng. Sci. (in press).

Bougrier, C., Delgenès, J.P., Carrère, H., 2008. Effects of thermal treatments on five different waste activated sludge samples solubilisation, physical properties and anaerobic digestion. Chem. Eng. J. 139, 236–244. https://doi.org/10.1016/j.cej.2007.07.099

Cao, Y., van Loosdrecht, M.C.M., Daigger, G.T., 2017. Mainstream partial nitritation–anammox in municipal wastewater treatment: status, bottlenecks, and further studies. Appl. Microbiol. Biotechnol. 101, 1365–1383. https://doi.org/10.1007/s00253-016-8058-7

Carballa, M., Fink, G., Omil, F., Lema, J.M., Ternes, T., 2008. Determination of the solid-water distribution coefficient (Kd) for pharmaceuticals, estrogens and musk fragrances in digested sludge. Water Res. 42, 287–295. https://doi.org/10.1016/j.watres.2007.07.012

Carballa, M., Omil, F., Lema, J.M., Llompart, M., García-Jares, C., Rodríguez, I., Gómez, M., Ternes, T., 2004. Behavior of pharmaceuticals, cosmetics and hormones in a sewage treatment plant. Water Res. 38, 2918–2926. https://doi.org/10.1016/j.watres.2004.03.029

Carballa, M., Omil, F., Ternes, T., Lema, J.M., 2007. Fate of pharmaceutical and personal care products (PPCPs) during anaerobic digestion of sewage sludge. Water Res. 41, 2139–2150. https://doi.org/10.1016/j.watres.2007.02.012

Clara, M., Gans, O., Windhofer, G., Krenn, U., Hartl, W., Braun, K., Scharf, S., Scheffknecht, C., 2011. Occurrence of polycyclic musks in wastewater and receiving water bodies and fate during wastewater treatment. Chemosphere 82, 1116–1123. https://doi.org/10.1016/j.chemosphere.2010.11.041

Collado, N., Buttiglieri, G., Ferrando-climent, L., Rodriguez-mozaz, S., Barceló, D., 2012. Removal of ibuprofen and its transformation products : Experimental and simulation studies. Sci. Total Environ. 433, 296–301. https://doi.org/10.1016/j.scitotenv.2012.06.060

Eberl, H., Morgenroth, E., Noguera, D., Picioreanu, C., Rittmann, B., van Loosdrecht, M., Wanner, O., 2006. Mathematical modeling of biofilms. IWA publishing London.

Fernadez-Fontaina, E., Omil, F., Lema, J.M., Carballa, M., 2012. Influence of nitrifying conditions on the biodegradation and sorption of emerging micropollutants. Water Res. 46, 5434–5444. https://doi.org/10.1016/j.watres.2012.07.037

Fernandez-Fontaina, E., Carballa, M., Omil, F., Lema, J.M., 2014. Modelling cometabolic biotransformation of organic micropollutants in nitrifying reactors. Water Res. 65, 371–383. https://doi.org/10.1016/j.watres.2014.07.048

Fernandez-Fontaina, E., Gomes, I.B., Aga, D.S., Omil, F., Lema, J.M., Carballa, M., 2016. Biotransformation of pharmaceuticals under nitrification, nitratation and heterotrophic conditions. Sci. Total Environ. 541, 1439–1447. https://doi.org/10.1016/j.scitotenv.2015.10.010

Fernandez-Fontaina, E., Pinho, I., Carballa, M., Omil, F., Lema, J.M., 2013. Biodegradation kinetic constants and sorption coefficients of micropollutants in membrane bioreactors. Biodegradation 24, 165–177. https://doi.org/10.1007/s10532-012-9568-3

Flores-Alsina, X., Arnell, M., Amerlinck, Y., Corominas, L., Gernaey, K. V., Guo, L., Lindblom, E., Nopens, I., Porro, J., Shaw, A., Snip, L., Vanrolleghem, P.A., Jeppsson, U., 2014. Balancing effluent quality, economic cost and greenhouse gas emissions during the evaluation of (plant-wide) control/operational strategies in WWTPs. Sci. Total Environ. 466–467, 616–624. https://doi.org/10.1016/j.scitotenv.2013.07.046

Flores-Alsina, X., Corominas, L., Snip, L., Vanrolleghem, P.A., 2011. Including greenhouse gas emissions during benchmarking of wastewater treatment plant control strategies. Water Res. 45, 4700–4710.

https://doi.org/10.1016/j.watres.2011.04.040

Gauthier, H., Yargeau, V., Cooper, D.G., 2010. Biodegradation of pharmaceuticals by Rhodococcus rhodochrous and Aspergillus niger by co-metabolism. Sci. Total Environ. 408, 1701–1706. https://doi.org/10.1016/j.scitotenv.2009.12.012

Gernaey, K.V., Jeppsson, U., Vanrolleghem, P.A., Copp, J.B., 2014. Benchmarking of control strategies for wastewater treatment plants. IWA Scientific and Technical Report no. 23. IWA Publishing. https://doi.org/https://doi.org/10.2166/9781780401171

Gernaey, K. V., Flores-Alsina, X., Rosen, C., Benedetti, L., Jeppsson, U., 2011. Dynamic influent pollutant disturbance scenario generation using a phenomenological modelling approach. Environ. Model. Softw. 26, 1255–1267. https://doi.org/10.1016/J.ENVSOFT.2011.06.001

Gikas, P., 2017. Towards energy positive wastewater treatment plants. J. Environ. Manage. 203, 621–629. https://doi.org/10.1016/j.jenvman.2016.05.061

Gonzalez-Gil, L., Mauricio-Iglesias, M., Carballa, M., Lema, J.M., 2018a. Why are organic micropollutants not fully biotransformed? A mechanistic modelling approach to anaerobic systems. Water Res. 142, 115–128. https://doi.org/https://doi.org/10.1016/j.watres.2018.05.032

Gonzalez-Gil, L., Mauricio-Iglesias, M., Serrano, D., Lema, J.M., Carballa, M., 2018b. Role of methanogenesis on the biotransformation of organic micropollutants during anaerobic digestion. Sci. Total Environ. 622–623, 459–466. https://doi.org/10.1016/j.scitotenv.2017.12.004

Gonzalez-Gil, L., Papa, M., Feretti, D., Ceretti, E., Mazzoleni, G., Steimberg, N., Pedrazzani, R., Bertanza, G., Lema, J.M., Carballa, M., 2016. Is anaerobic digestion effective for the removal of organic micropollutants and biological activities from sewage sludge? Water Res. 102, 211–220. https://doi.org/10.1016/j.watres.2016.06.025

Gu, Y., Li, Y., Li, X., Luo, P., Wang, H., Robinson, Z.P., Wang, X., Wu, J., Li, F., 2017. The feasibility and challenges of energy self-sufficient wastewater treatment plants. Appl. Energy 204, 1463–1475. https://doi.org/10.1016/j.apenergy.2017.02.069

Guo, L., Vanrolleghem, P.A., 2014. Calibration and validation of an activated sludge model for greenhouse gases no. 1 (ASMG1): Prediction of temperature-dependent N2O emission dynamics. Bioprocess Biosyst. Eng. 37, 151–163. https://doi.org/10.1007/s00449-013-0978-3

Helton, J.C., Davis, F.J., 2003. Latin hypercube sampling and the propagation of uncertainty in analyses of complex systems. Reliab. Eng. Syst. Saf. 81, 23–69. https://doi.org/10.1016/S0951-8320(03)00058-9

Hyland, K.C., Dickenson, E.R.V., Drewes, J.E., Higgins, C.P., 2012. Sorption of ionized and neutral emerging trace organic compounds onto activated sludge from different wastewater treatment configurations. Water Res. 46, 1958–1968. https://doi.org/10.1016/j.watres.2012.01.012

Jimenez, J., Miller, M., Bott, C., Murthy, S., De Clippeleir, H., Wett, B., 2015. High-rate activated sludge system for carbon management - Evaluation of crucial process mechanisms and design parameters. Water Res. 87, 476–482. https://doi.org/10.1016/j.watres.2015.07.032

Jimenez, J.A., La Motta, E.J., Parker, D.S., 2005. Kinetics of Removal of Particulate Chemical Oxygen Demand in the Activated-Sludge Process. Water Environ. Res. 77, 437–446. https://doi.org/10.2175/106143005X67340

Jones, O.A.H., Voulvoulis, N., Lester, J.N., 2002. Aquatic environmental assessment of the top 25 English prescription pharmaceuticals. Water Res. 36, 5013–5022. https://doi.org/10.1016/S0043-1354(02)00227-0

Joss, A., Andersen, H., Ternes, T., Richle, P.R., Siegrist, H., 2004. Removal of estrogens in municipal wastewater treatment under aerobic and anaerobic conditions: Consequences for plant optimization. Environ. Sci. Technol. 38, 3047–3055. https://doi.org/10.1021/es0351488

Joss, A., Keller, E., Alder, A.C., Göbel, A., McArdell, C.S., Ternes, T., Siegrist, H., 2005. Removal of pharmaceuticals and fragrances in biological wastewater treatment. Water Res. 39, 3139–3152. https://doi.org/10.1016/j.watres.2005.05.031

Joss, A., Zabczynski, S., Go, A., Hoffmann, B., Lo, D., Mcardell, C.S., Ternes, T.A., Thomsen, A., Siegrist, H., 2006. Biological degradation of pharmaceuticals in municipal wastewater treatment : Proposing a classification scheme. Water Res. 40, 1686–1696. https://doi.org/10.1016/j.watres.2006.02.014

Kassotaki, E., Buttiglieri, G., Ferrando-Climent, L., Rodriguez-Roda, I., Pijuan, M., 2016. Enhanced sulfamethoxazole degradation through ammonia oxidizing bacteria co-metabolism and fate of transformation products. Water Res. 94, 111–119. https://doi.org/10.1016/j.watres.2016.02.022

Kassotaki, E., Pijuan, M., Joss, A., Borrego, C.M., Rodriguez-Roda, I., Buttiglieri, G., 2018. Unraveling the potential of a combined nitritation-anammox biomass towards the biodegradation of pharmaceutically active compounds. Sci. Total Environ. 624, 722–731. https://doi.org/10.1016/j.scitotenv.2017.12.116

Laureni, M., Falås, P., Robin, O., Wick, A., Weissbrodt, D.G., Nielsen, J.L., Ternes, T.A., Morgenroth, E., Joss, A., 2016. Mainstream partial nitritation and anammox: Long-term process stability and effluent quality at low temperatures. Water Res. 101, 628–639. https://doi.org/10.1016/j.watres.2016.05.005

Lautz, L.S., Struijs, J., Nolte, T.M., Breure, A.M., van der Grinten, E., van de Meent, D., van Zelm, R., 2017. Evaluation of SimpleTreat 4.0: Simulations of pharmaceutical removal in wastewater treatment plant facilities. Chemosphere 168, 870–876. https://doi.org/10.1016/j.chemosphere.2016.10.123

Lindblom, E., Arnell, M., Flores-Alsina, X., Stenström, F., Gustavsson, D.J.I., Yang, J., Jeppsson, U., 2016. Dynamic modelling of nitrous oxide emissions from three Swedish sludge liquor treatment systems. Water Sci. Technol. 73, 798–806.

Lotti, T., Kleerebezem, R., Hu, Z., Kartal, B., de Kreuk, M.K., van Erp Taalman Kip, C., Kruit, J., Hendrickx, T.L.G., van Loosdrecht, M.C.M., 2014. Pilot-scale evaluation of anammox-based mainstream nitrogen removal from municipal wastewater. Environ. Technol. 36, 1167–77. https://doi.org/10.1080/09593330.2014.982722

Luo, Y., Guo, W., Ngo, H.H., Nghiem, L.D., Hai, F.I., Zhang, J., Liang, S., Wang, X.C., 2014. A review on the occurrence of micropollutants in the aquatic environment and their fate and removal during wastewater treatment. Sci. Total Environ. 473–474, 619–641. https://doi.org/10.1016/j.scitotenv.2013.12.065

Malmborg, J., Magnér, J., 2015. Pharmaceutical residues in sewage sludge: Effect of sanitization and anaerobic digestion. J. Environ. Manage. 153, 1–10. https://doi.org/10.1016/j.jenvman.2015.01.041

Malovanyy, A., Trela, J., Plaza, E., 2015. Mainstream wastewater treatment in integrated fixed film activated sludge (IFAS) reactor by partial nitritation/anammox process. Bioresour. Technol. 198, 478–487.

Mbamba, C.K., Lindblom, E., Flores-Alsina, X., Tait, S., Anderson, S., Saagi, R., Batstone, D.J., Gernaey, K.V., Jeppsson, U., 2019. Plant-wide model-based analysis of iron dosage strategies for chemical phosphorus removal in wastewater treatment systems. Water Res. https://doi.org/10.1016/J.WATRES.2019.01.048

Men, Y., Achermann, S., Helbling, D.E., Johnson, D.R., Fenner, K., 2017. Relative contribution of ammonia oxidizing bacteria and other members of nitrifying activated sludge communities to micropollutant biotransformation. Water Res. 109, 217–226. https://doi.org/10.1016/j.watres.2016.11.048

Narumiya, M., Nakada, N., Yamashita, N., Tanaka, H., 2013. Phase distribution and removal of pharmaceuticals and personal care products during anaerobic sludge digestion. J. Hazard. Mater. 260, 305–312. https://doi.org/10.1016/j.jhazmat.2013.05.032

Paterakis, N., Chiu, T.Y., Koh, Y.K.K., Lester, J.N., McAdam, E.J., Scrimshaw, M.D., Soares, A., Cartmell, E., 2012. The effectiveness of anaerobic digestion in removing estrogens and nonylphenol ethoxylates. J. Hazard. Mater. 199–200, 88–95. https://doi.org/10.1016/j.jhazmat.2011.10.075

Polesel, F., Andersen, H.R., Trapp, S., Plosz, B.G., 2016. Removal of Antibiotics in Biological Wastewater Treatment Systems□ A Critical Assessment Using the Activated Sludge Modeling Framework for Xenobiotics (ASM-X). Environ. Sci. Technol. 50, 10316–10334.

Pomiès, M., Choubert, J.M., Wisniewski, C., Coquery, M., 2013. Modelling of micropollutant removal in biological wastewater treatments: A review. Sci. Total Environ. 443, 733–748. https://doi.org/10.1016/j.scitotenv.2012.11.037

Radjenović, J., Petrović, M., Barceló, D., 2009. Fate and distribution of pharmaceuticals in wastewater and sewage sludge of the conventional activated sludge (CAS) and advanced membrane bioreactor (MBR) treatment. Water Res. 43, 831–841. https://doi.org/10.1016/j.watres.2008.11.043

Reyes-Contreras, C., Neumann, P., Barriga, F., Venegas, M., Domínguez, C., Bayona, J.M., Vidal, G., 2018. Organic micropollutants in sewage sludge: influence of thermal and ultrasound hydrolysis processes prior to anaerobic stabilization. Environ. Technol. 1–22. https://doi.org/10.1080/09593330.2018.1534892

Saltelli, A., Ratto, M., Andres, T., Campolongo, F., Cariboni, J., Gatelli, D., Saisana, M., Tarantola, S., 2008. GLOBAL SENSITIVITY ANALYSIS. The global. West Sussex.

Samaras, V.G., Stasinakis, A.S., Thomaidis, N.S., Mamais, D., Lekkas, T.D., 2014. Fate of selected emerging micropollutants during mesophilic, thermophilic and temperature co-phased anaerobic digestion of sewage sludge. Bioresour. Technol. 162, 365–372. https://doi.org/10.1016/j.biortech.2014.03.154

Santos, J.L., Aparicio, I., Callejón, M., Alonso, E., 2009. Occurrence of pharmaceutically active compounds during 1-year period in wastewaters from four wastewater treatment plants in Seville (Spain). J. Hazard. Mater. 164, 1509–1516. https://doi.org/10.1016/j.jhazmat.2008.09.073

Sapkaite, I., Barrado, E., Fdz-Polanco, F., Pérez-Elvira, S.I., 2017. Optimization of a thermal hydrolysis process for sludge pre-treatment. J. Environ. Manage. 192, 25–30. https://doi.org/10.1016/j.jenvman.2017.01.043

Sin, G., Gernaey, K. V, Lantz, A.E., 2009. Good modeling practice for PAT applications: Propagation of input uncertainty and sensitivity analysis. Biotechnol. Prog. 25, 1043–1053. https://doi.org/10.1002/btpr.166

Smitshuijzen, J., Pérez, J., Duin, O., Loosdrecht, M.C.M. va., 2016. A simple model to describe the performance of highly-loaded aerobic COD removal reactors. Biochem. Eng. J. 112, 94–102. https://doi.org/10.1016/J.BEJ.2016.04.004

Snip, L.J.P., Flores-Alsina, X., Plósz, B.G., Jeppsson, U., Gernaey, K. V, 2014. Modelling the occurrence, transport and fate of pharmaceuticals in wastewater systems. Environ. Model. Softw. 62, 112–127.

Stasinakis, A.S., Thomaidis, N.S., Arvaniti, O.S., Asimakopoulos, A.G., Samaras, V.G., Ajibola, A., Mamais, D., Lekkas, T.D., 2013. Contribution of primary and secondary treatment on the removal of benzothiazoles, benzotriazoles, endocrine disruptors, pharmaceuticals and perfluorinated compounds in a sewage treatment plant. Sci. Total Environ. 463–464, 1067–1075. https://doi.org/10.1016/j.scitotenv.2013.06.087

Stevens-Garmon, J., Drewes, E., Khan, S.J., Mcdonald, J.A., Dickenson, E.R. V, 2011. Sorption of emerging trace organic compounds onto wastewater sludge solids. Water Res. 45, 3417–3426. https://doi.org/10.1016/j.watres.2011.03.056

Struijs, J., van de Meent, D., Schowanek, D., Buchholz, H., Patoux, R., Wolf, T., Austin, T., Tolls, J., van Leeuwen, K., Galay-Burgos, M., 2016. Adapting SimpleTreat for simulating behaviour of chemical substances during industrial sewage treatment. Chemosphere 159, 619–627. https://doi.org/10.1016/j.chemosphere.2016.06.063

Suárez, S., Carballa, M., Omil, F., Lema, J.M., 2008. How are pharmaceutical and personal care products (PPCPs) removed from urban wastewaters? Rev. Environ. Sci. Biotechnol. 7, 125–138. https://doi.org/10.1007/s11157-008-9130-2

Suarez, S., Lema, J.M., Omil, F., 2010. Removal of Pharmaceutical and Personal Care Products (PPCPs) under nitrifying and denitrifying conditions. Water Res. 44, 3214–3224. https://doi.org/10.1016/j.watres.2010.02.040

Taboada-Santos, A., Braz, G.H.R., Fernandez-Gonzalez, N., Carballa, M., Lema, J.M., 2019a. Thermal hydrolysis of sewage sludge partially removes organic micropollutants but does not enhance their anaerobic biotransformation. Sci. Total Environ. 690, 534–542. https://doi.org/10.1016/j.scitotenv.2019.06.492

Taboada-Santos, A., Lema, J.M., Carballa, M., 2019b. Opportunities for rotating belt filters in novel wastewater treatment plant configurations. Environ. Sci. Water Res. Technol. 5, 704–712. https://doi.org/10.1039/C8EW00899J

Taboada-Santos, A., Lema, J.M., Carballa, M., 2019c. Energetic and economic assessment of sludge thermal hydrolysis in novel wastewater treatment plant configurations. Waste Manag. 92, 30–38.

Taboada-Santos, A., Rivadulla, E., Paredes, L., Carballa, M., Romalde, J.L., Lema, J.M., 2020. Comprehensive comparative between chemically enhanced primary treatment and high-rate activated sludge for preconcentration of organic carbon in novel wastewater treatment plant configurations. Water Res. 169, 115-258.

Takács, I., Patry, G.G., Nolasco, D., 1991. A dynamic model of the clarification-thickening process. Water Res. 25, 1263–1271. https://doi.org/10.1016/0043-1354(91)90066-Y

Ternes, T.A., Herrmann, N., Bonerz, M., Knacker, T., Siegrist, H., Joss, A., 2004. A rapid method to measure the solid-water distribution coefficient (Kd) for pharmaceuticals and musk fragrances in sewage sludge. Water Res. 38, 4075–4084. https://doi.org/10.1016/j.watres.2004.07.015

Vangsgaard, A.K., Mauricio-Iglesias, M., Gernaey, K. V., Smets, B.F., Sin, G., 2012. Sensitivity analysis of autotrophic N removal by a granule based bioreactor: Influence of mass transfer versus microbial kinetics. Bioresour. Technol. 123, 230–241. https://doi.org/10.1016/j.biortech.2012.07.087

Vangsgaard, A.K., Mutlu, A.G., Gernaey, K. V, Smets, B.F., Sin, G., 2013. Calibration and validation of a model describing complete autotrophic nitrogen removal in a granular SBR system. J. Chem. Technol. Biotechnol. 88, 2007–2015.

Verlicchi, P., Al Aukidy, M., Zambello, E., 2012. Occurrence of pharmaceutical compounds in urban wastewater: Removal, mass load and environmental risk after a secondary treatment-A review. Sci. Total Environ. 429, 123–155. https://doi.org/10.1016/j.scitotenv.2012.04.028

Wan, J., Gu, J., Zhao, Q., Liu, Y., 2016. COD capture: A feasible option towards energy self-sufficient domestic wastewater treatment. Sci. Rep. 6, 1–9. https://doi.org/10.1038/srep25054

Wick, A., Fink, G., Joss, A., Siegrist, H., Ternes, T.A., 2009. Fate of beta blockers and psycho-active drugs in conventional wastewater treatment. Water Res. 43, 1060–1074. https://doi.org/10.1016/j.watres.2008.11.031

Xue, W., Wu, C., Xiao, K., Huang, X., Zhou, H., 2010. Elimination and fate of selected micro-organic pollutants in a full-scale anaerobic / anoxic / aerobic process combined with membrane bioreactor for municipal wastewater reclamation. Water Res. 44, 5999–6010. https://doi.org/10.1016/j.watres.2010.07.052

Yang, S., Hai, F.I., Price, W.E., McDonald, J., Khan, S.J., Nghiem, L.D., 2016. Occurrence of trace organic contaminants in wastewater sludge and their removals by anaerobic digestion. Bioresour. Technol. 210, 153–159. https://doi.org/10.1016/j.biortech.2015.12.080

Zhang, X., Li, R., 2018. Variation of antibiotics in sludge pretreatment and anaerobic digestion processes: Degradation and solid-liquid distribution. Bioresour. Technol. 255, 266–272. https://doi.org/10.1016/j.biortech.2018.01.100

Table legends

Table 1. Average and standard deviation of the solid-liquid distribution constants (K_D) values (in bold) considered in this study and range of the values found in the literature.

Table 2. Average and standard deviation of the pseudo first-order biotransformation constants (k_{biol}) (in bold) considered in this study and range of the values found in the literature.

Table 3. Average and standard deviation of the OMP removal efficiency in AD (in bold) and range of the values found in the literature.

Table 4. Comparison of energy requirements, digested sludge production, methane production and effluent quality in novel and conventional WWTP configurations.

Table 5. Presence of the representative OMPs of each group in the WWTPs effluents and in digested sludge.

Table 1.

OMP	K_D (L/kg TSS)						
	Influent	CEPT sludge	HRAS sludge	RBF sludge	Primary sludge	CAS sludge	Anaerobic sludge
AHTN	**8,857 ± 2,148** [1]	**5,286 ± 1,066** [1]	**9,969 ± 2,557** [1]	**24,247 ± 6,069** [2]	**5,300 ± 205** 5,300 ± 1,900 [3] 5,010 [4]	**4,200 ± 1,356** 2,400 ± 960 [3] 6000 ± 300 [5] 2,714 ± 1,313 [6] 2,571-2,838 [7] 3,347 ± 1,900 [8]	**14,050 ± 6,082** 3,000 ± 2,000 [5] 11,375 [9] 15,200 ± 7,800 [10] 16,500 - 72,000 [11]
ABDI	**3,856 ± 845** [1]	**2,461 ± 411** [1]	**4,574 ± 832** [1]	**12,003 ± 4,037** [2]	**5,010 ± 0** [4]	**5,142 ± 2,531** [6]	**1,200 ± 500** [5]
HHCB	**5,927 ± 2,168** [1]	**3,412 ± 679** [1]	**6,853 ± 1,945** [1]	**57,450 ± 9,770** [2]	**4,920 ± 64** 4,920 ± 2,080 [3] 5,010 [4]	**2,110 ± 396** 1,616 ± 772 [6] 2,214-2,478 [7] 2428 ± 1297 [8]	**9,700 ± 5,208** 3,700 ± 1,200 [5] 12,000 [9] 13,300 ± 5,500 [10]
TCS	**10,439 ± 2,170** [1]	**5,918 ± 225** [1]	**8,748 ± 1,635** [1]	**27,947 ± 18,228** [2]	**3,650 ± 1,230** 1,000-6,310*[12] *Mixed sludge	**5,725 ± 2,703** 1,905-9,549 [13]	**7,020 ± 1,519** 3,630-22,390 [11] 794-1,259 [12]
IBP	**8 ± 8** [1]	**15 ± 2** [1]	**16 ± 16** [1]	**92 ± 25** [2]	**225 ± 217** <20 [3] 9.5 ± 3.1 [14] 453 [15] <30 [16]	**210 ± 139** 7 ± 2 [3] 240 ± 10 [5] 24 ± 5 [6] 33-80 [7] <30 [16] 144-417 [13] 6 ± 4 [17]	**60 ± 29** 100 ± 100 [5] 31 [9] 38 ± 14 [10] 11-58 [11] 20-40 [12]
NPX	**9 ± 9** [1]	**0 ± 0** [1]	**18 ± 12** [1]	**1 ± 1** [2]	**125 ± 125** 217 [15] <30 [16]	**125 ± 86** 100 ± 10 [5] 17 ± 6 [6] 36-58 [7] 217 [15] <30 [16] 79-245 [13] 10 ± 1 [17]	**10 ± 6** 0 [5] <50 [9] 11 [10] 0 [11]

Table 1 *(cont.)*.

OMP	K_D (L/kg SS)						
	Influent	CEPT sludge	HRAS sludge	Cellulosic sludge	Primary sludge	CAS sludge	Anaerobic sludge
DCF	**13 ± 3** [1]	**7 ± 5** [1]	**21 ± 2** [1] [1]	**10 ± 8** [2]	**245 ± 207** 459 ± 32 [3] 500 [10] 194 ± 134 [14] 459 ± 210 [18] <30 [16]	**155 ± 110** 16 ± 3 [3] 0 [5] 32 ± 14 [6] <6 [7] 120 [14] 232 ± 139 [18] <30 [16] 81-309 [13]	**79 ± 6** 0 [5] 600 [9] 66 ± 23 [10] 79-158 [12]
ERY	**25 ± 10** [1]	**87 ± 18** [1]	**40 ± 11** [1]	**6 ± 4** [2]	**235 ± 102** 309 ± 272 [14] 165 [15]	**50 ± 19** 50 ± 10 [5] 28 ± 10 [6] 49-70 [7] 74 ± 26 [8]	**630 ± 557** 30 ± 15 [5] 40-1,260 [12]
ROX	**54 ± 13** [1]	**200 ± 25** [1]	**69 ± 12** [1]	**14 ± 5** [2]	**400 ± 0** 400 [19]	**296 ± 183** 100 ± 10 [5] 51 ± 11 [6] 80-99 [7] 75 ± 48 [8] 170 [19] 570 ± 60 [17]	**1,000 ± 982** 40 ± 30 [5] 2,000 [9] 83 [10] 80-2,000 [12]
SMX	**35 ± 11** [1]	**45 ± 15** [1]	**52 ± 20** [1]	**14 ± 2** [2]	**15 ± 19** 3.2 ± 4.5 [14] <30 [16]	**50 ± 28** 80 ± 10 [5] 11 ± 7 [6] 33-63 [7] <30 [16] 50 ± 13 [17] 87-851[13]	**250 ± 212** 45 ± 30 [5] 500 [9] 23 [10] 16-25 [12]

 Table 1 *(cont.)*.

OMP	**K_D (L/kg SS)**						
	Influent	CEPT sludge	HRAS sludge	Cellulosic sludge	Primary sludge	CAS sludge	Anaerobic sludge
TMP	**162 ± 42** [1]	**108 ± 18** [1]	**188** **188 ± 28** [1]	**48 ± 2** [2]	**339 ± 124** 427 ± 238 [14] 251 ± 99 [16]	**212 ± 125** 80 ± 5 [5] 25 ± 8 [6] 61-90 [7] 45 ± 30 [8] 119 ± 49 [16] 193 ± 104 [16] 178-398 [13] 330 ± 25 [17]	**368 ± 335** 12 ± 7 [5] 83 - 724 [11]
FLX	**1,420 ± 124** [1]	**1,518 ± 219** [1]	**1,7501,750 ± 575** [1]	**228 ± 37** [2]	**639 ± 69** 590-687 [11]	**1,430 ± 798** 2,500 ± 200 [5] 355 ± 145 [6] 762-1,043 [7] 1,603 ± 905 [8] 851-1,820 [13]	**1,515 ± 1326** 700 ± 200 [5] 275-2,754 [11]
CBZ	**53 ± 15** [1]	**101 ± 21** [1]	**76 ± 15** [1]	**17 ± 13** [2]	**167 ± 131** <20 [3] 314 ± 205 [14] 178 [15] 65 ± 5 [16]	**117 ± 69** 1.2 ± 0.5 [3] 0 [5] <1.0 [6] 15-20 [7] 135 [14] 36 ± 2 [16] 50 ± 1 [16] 47-234 [13] <75 [17] 17 ± 1 [20]	**300 ± 74** 0 [5] 20 [9] 35 [10] 40 - 186 [11]

Table 1 *(cont.).*

OMP	K_D (L/kg SS)						
	Influent	CEPT sludge	HRAS sludge	Cellulosic sludge	Primary sludge	CAS sludge	Anaerobic sludge
DZP	**95 ± 17** [1]	**141 ± 22** [1]	**166 ± 14** [1]	**237 ± 123** [2]	**168 ± 160** 44 ± 26[3] 291 ± 50 [16]	**131 ± 91** 21 ± 8 [3] 30 ± 10 [5] 50 ± 14 [6] 78-137 [7] 116 ± 52 [8] 197 ± 31 [16] 241 ± 59 [16] 81-295 [13] 53 ± 1 [20]	**290 ± 179** 400 ± 250 [5] 0 [9] 71 - 76 [11]
E1	**399 ± 49** [1]	**322 ± 22** [1]	**346 ± 150** [1]	**131 ± 8** [2]	**636 ± 104** 636 ± 104 [16]	**373 ± 290** 150 ± 30 [5] 607 ± 48 [16] 645 ± 87 [16] <100 [17]	**235 ± 162** 300 ± 250 [5] <250 [9] 303 ± 59 [10] 58 - 813 [11]
E2	**359 ± 53** [1]	**265 ± 23** [1]	**599 ± 19** [1]	**132 ± 39** [2]	**560 ± 67** 560 ± 67 [16]	**667 ± 147** 800 ± 100 [5] 771 ± 108 [16] 533 ± 34 [16]	**436 ± 152** 250 ± 150 [5] <1,000 [9] 461 ± 212 [10] 166 - 2,188 [11]
EE2	**529 ± 44** [1]	**407 ± 26** [1]	**464 ± 6** [1]	**76 ± 23** [2]	**634 ± 435** 278 ± 3 [3] 251 [10] 1,017 ± 105 [16]	**875 ± 539** 349 ± 4 7 [3] 200 ± 100 [5] 1,103 ± 76 [16] 1,550 ± 223 [16] 300-500 [17]	**224 ± 207** 300 ± 250 [5] <1,000 [9] 432 ± 168 [10] 16-25 [11]

1. Taboada-Santos et al., (2020), 2. Taboada-Santos et al. (2019b), 3. Ternes et al. (2004), 4. Suárez et al. (2008), 5. Alvarino et al. (2014), 6. Fernandez-Fontaina et al. (2013), 7. Fernandez-Fontaina et al. (2014), 8. Fernandez-Fontaina et al. (2012), 9. Carballa et al. (2007), 10. Carballa et al. (2008), 11. Gonzalez-Gil et al. (2016), 12. Narumiya et al. (2013), 13. Hyland et al. (2012), 14. Radjenović et al. (2009), 15. Jones et al. (2002), 16. Stevens-Garmon et al. (2011), 17. Abegglen et al. (2009), 18. Wick et al. (2009), 19. Joss et al. (2005), 20. Wick et al. (2009).

Table 2.

OMP	k_{biol} (L/g $_{VSS}$ ·d) HRAS sludge	PN-AMX sludge	CAS sludge
AHTN	**15 ± 5** [1]	**0.5 ± 0.3** [21]	**60 ± 43**
			38 ± 16 [5]
			2 ± 2 [5]
			3.9 [6]
			14.2 [7]
			15.7 [7]
			115 [22]
			0.02 [23]
ADBI	**16 ± 4** [1]	**1.3 ± 0.6** [21]	**40 ± 36**
			6 ± 1 [5]
			63 ± 25 [5]
			9.1 [6]
			75 [22]
HHCB	**11 ± 4** [1]	**0.8 ± 0.3** [21]	**30 ± 17**
			7 ± 1 [5]
			41 ± 42 [5]
			1.7 [6]
			20.9 [7]
			32.9 [7]
			170 [22]
TCS	**13 ± 5** [1]	**0.7 ± 0.3** [21]	**0.7 ± 0.3** [1]
IBP	**29 ± 9** [1]	**38 ± 7** [21]	**20 ± 12**
			2 ± 1 [5]
			24 ± 7 [5]
			6 [6]
			2.4 [7]
			4.3 [7]
			1.3-3 [17]
			20 [22]
			21-35 [23]
NPX	**7 ± 3** [1]	**17 ± 0** [21]	**5.0 ± 3.6**
			1 ± 0 [5]
			9 ± 2 [5]
			0.5 [6]
			1.4 [7]
			2.6 [7]
			0.1 [17]
			9 [22]
			1-1.9 [23]
DCF	**0.5 ± 0.4** [1]	**0.9 ± 0.1** [21]	**0.05 ± 0.04**
			2 ± 1 [5]
			0.1 ± 0.1 [5]
			0.02 [6]
			0 [7]
			<0.02 [17]
			1.2 [22]
			0.03-0.05 [23]

Table 2 *(cont.)*.

OMP	k_{biol} (L/g $_{VSS}$ ·d) HRAS sludge	PN-AMX sludge	CAS sludge
ERY	**0.9 ± 0.8** [1]	**0.5 ± 0.3** [21]	**3.0 ± 2.1**
			1 ± 1 [5]
			3 ± 0 [5]
			0.5 [6]
			0.8 [7]
			3 [7]
			6 [22]
			0.1 [23]
ROX	**3.3 ± 2.1** [1]	**0.3 ± 0.1** [21]	**5.0 ± 3.4**
			8 ± 3 [5]
			2.2 ± 1.5 [5]
			1.2 [6]
			2.3-3.4 [7]
			0.023 ± 0.018 [17]
			9 [22]
			0.1 [23]
SMX	**1.5 ± 0.8** [1]	**0.3 ± 0.1** [21]	**4.5 ± 3.6**
			0.7 ± 0.9 [5]
			9 ± 1 [5]
			0.1 [6]
			1 [7]
			0.3 [7]
			0.2 [17]
			0.3 [22]
			5.9-7.6 [23]
TMP	**0.4 ± 0.3** [1]	**0.2 ± 0.1** [21]	**0.3 ± 0.2**
			0.6 ± 0.3 [5]
			0 [5]
			0.09 [6]
			0 [7]
			0.9 [7]
			0.22 ± 0.02 [17]
			0.15 [22]
FLX	**1.3 ± 1.1** [1]	**0.10 ± 0.05** [21]	**6.0 ± 4.3**
			10 ± 1 [5]
			0.8 ± 0.5 [5]
			1.98 [6]
			0.6 [7]
			1.3 [7]
			9 [22]
CBZ	**0.4 ± 0.3** [1]	**0 ± 0** [21]	**0.1 ± 0.1**
			0.2 ± 0.1 [5]
			0 [5]
			0.01 [6]
			0 [7]
			<0.008 [17]
			<0.1 [20]
			0.1 [22]
			<0.01 [23]

Table 2 *(cont.)*.

OMP	k_{biol} (L/g$_{VSS}$·d) HRAS sludge	PN-AMX sludge	CAS sludge
DZP	**2.6 ± 1.6** [1]	**0 ± 0** [21]	**0.2 ± 0.2**
			0.4 ± 0.1 [5]
			0.02 ± 0.00 [5]
			0.19 [6]
			0 [7]
			<0.16 [20]
			0.4 [22]
			0.02-0.04 [23]
E1	**57 ± 20** [1]	**53 ± 14** [21]	**85 ± 79**
			2 ± 4 [5]
			14 ± 13 [5]
			170 [22]
			>100 [23]
			162 ± 25 [24]
E2	**46 ± 10** [1]	**27 ± 12** [21]	**180 ± 149**
			19 ± 14 [5]
			11 ± 13 [5]
			170 [22]
			>100[23]
			350 ± 42 [24]
EE2	**1.7 ± 0.9** [1]	**2 ± 1** [21]	**10 ± 1**
			7 ± 4 [5]
			2 ± 1 [5]
			20 [22]
			5-10 [23]
			8 ± 2 [24]

1. Taboada-Santos et al. (2020), 5. Alvarino et al. (2014), 6. Fernandez-Fontaina et al. (2013), 7. Fernandez-Fontaina et al. (2014), 17. Abegglen et al. (2009), 20. Wick et al. (2009), 21. Alvarino et al. (2015), 22. Suarez et al. (2010), 23. Joss et al. (2006), 24. Joss et al. (2004).

Table 3.

OMP	Biotransformation during AD
AHTN	**30 ± 25**
	60 [9]
	0 [11]
	0/45 [25]
	40 [26]
	30/60 [27]
ABDI	**30 ± 0 ***
HHCB	**30 ± 23**
	60 [9]
	10 [11]
	40 [26]
	50/70 [27]
TCS	**40 ± 18**
	20 [11]
	30 [12]
	50 [26]
	65 [28]
	50 [29]
IBP	**45 ± 29**
	45 [9]
	30 [11]
	25 [26]
	70-75 [27]
	95 [28]
	10 [29]
	30 [30]
NPX	**90 ± 13**
	85 [9]
	100 [11]
	60 [26]
	100 [27]
	85 [28]
	90 [29]
	85 [30]
DCF	**50 ± 38**
	0/80 [9]
	25 [12]
	20 [26]
	95 [28]
	25 [30]
ERY	**45 ± 7**
	45 [12]
	35 [26]
ROX	**50 ± 34**
	95 [9]
	85 [11]
	65 [12]
	25 [26]
	65/70 [27]
	0 [29]
SMX	**95 ± 5**
	100 [9]
	80 [11]
	100 [12]
	100 [26]

Table 3 *(cont.)*.

OMP	Biotransformation during AD
TMP	**70 ± 27**
	75 [11]
	100 [12]
	75 [26]
	35/40 [27]
	90 [29]
	100 [30]
FLX	**35 ± 22**
	70 [11]
	35 [26]
	25-30 [27]
	30 [29]
	0 [30]
	30 [31]
CBZ	**20 ± 10**
	5 [9]
	30 [11]
	0 [12]
	10 [26]
	10/20 [27]
	15 [30]
	0 [29]
DZP	**55 ± 20**
	30 [9]
	50 [11]
	35 [26]
	70/75 [27]
E1+E2	**40 ± 31**
	80 [9]
	0 [11]
	35 [26]
	0/10 [27]
	0 [30]
	50 [32]
EE2	**50 ± 35**
	40/95 [9]
	75 [11]
	45 [26]
	0 [30]
	20 [32]

9. Carballa et al. (2007), 11. Gonzalez-Gil et al. (2016), 12. Narumiya et al. (2013), 25. Clara et al. (2011), 26. Gonzalez-Gil et al. (2018), 27. Taboada-Santos et al. (2019a), 28. Samaras et al. (2014), 29. Yang et al. (2016), 30. Malmborg and Magnér (2015), 31. Bergersen et al. (2012), 32. Paterakis et al. (2012).

* Assumed as the one of other musk fragrances.

Table 4.

Parameters	HRAS	RBF+HRAS	CEPT	Conventional
Aeration demand (kWh/d)	1,997	1,881	1,311	4,216
CH_4 production (Nm^3/d)	2,161	2,295	2,351	1,719
Digested sludge (ton TS/d)	2.7	2.6	3.6	2.3
Effluent COD (g COD/m^3)	46	45	28	47
Effluent TN (g N/m^3)	4.4	3.8	4.2	17.6

Table 5

Stream	TCS	IBP	EE2	CBZ
Influent WWTP (mg/d)	206	206	20.6	206
Effluent HRAS (mg/d)	26	2.5	8.2	191
Effluent RBF+HRAS (mg/d)	25	3.6	8.6	195
Effluent CEPT (mg/d)	34	12	9.3	202
Effluent conventional (mg/d)	46	6	1.0	172
Digested sludge HRAS (mg/d)	36	0.1	0.8	3.5
Digested sludge RBF+HRAS (mg/d)	48	0.2	0.8	3.1
Digested sludge CEPT (mg/d)	78	0.4	1.1	3.0
Digested sludge conventional (mg/d)	74	0.5	0.7	3.7

Figure captions

Figure 1. Novel and conventional WWTP configurations considered for assessing the fate of OMPs.

Figure 2. OMPs removal efficiency from wastewater achieved in the HRAS-based configuration ( ), the RBF+HRAS configuration ( ), the CEPT-based configuration ( ) and the conventional configuration ( ).

Figure 3. Presence of OMPs in digested sludge in the HRAS-based configuration ( ), the RBF+HRAS configuration ( ), the CEPT-based configuration ( ) and the conventional configuration ( ).

Figure 1.

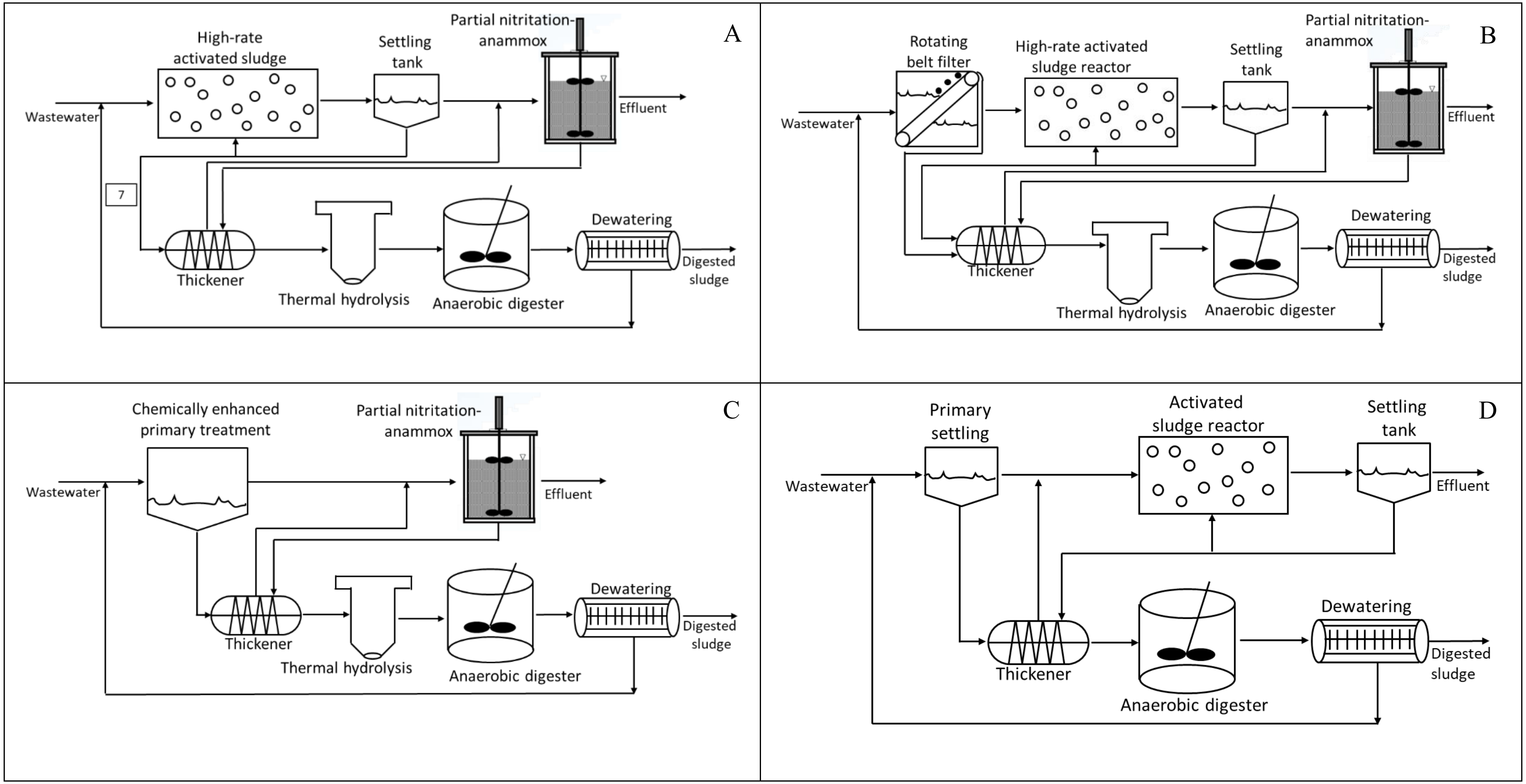
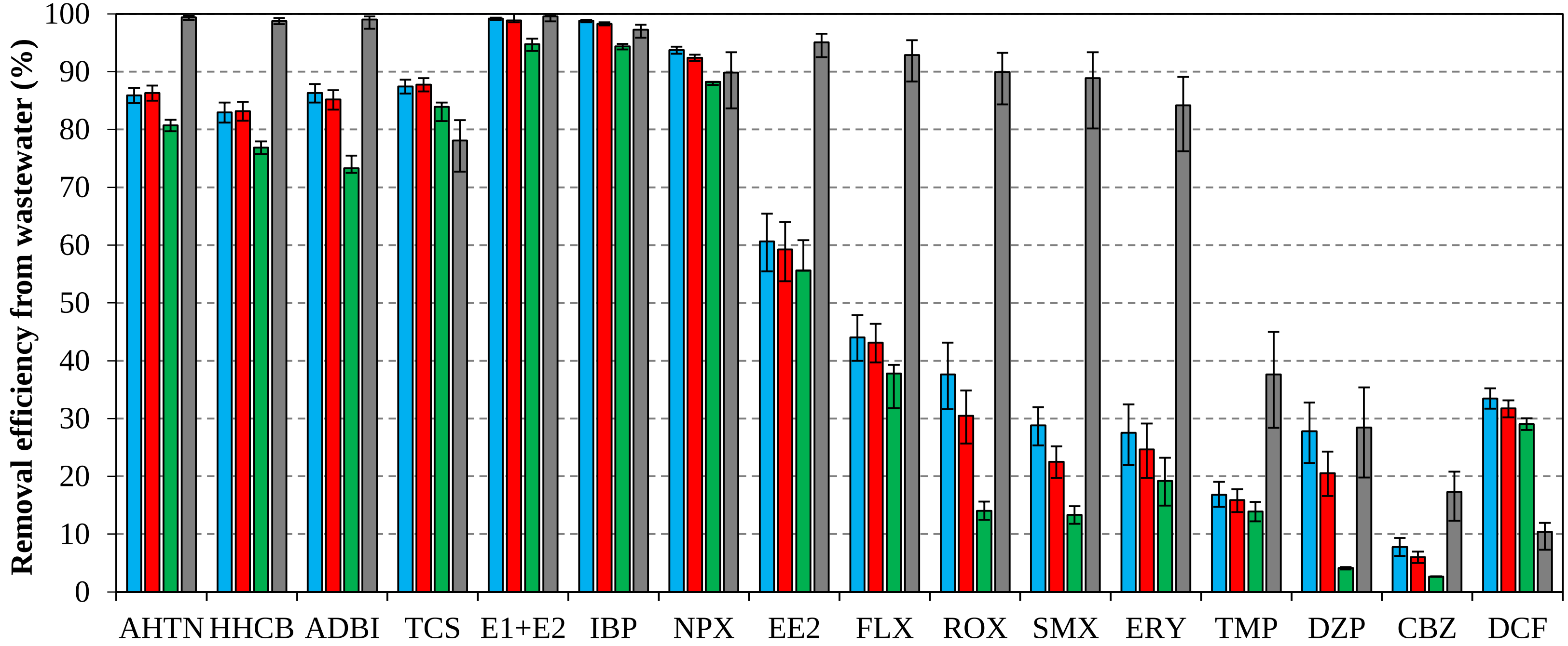
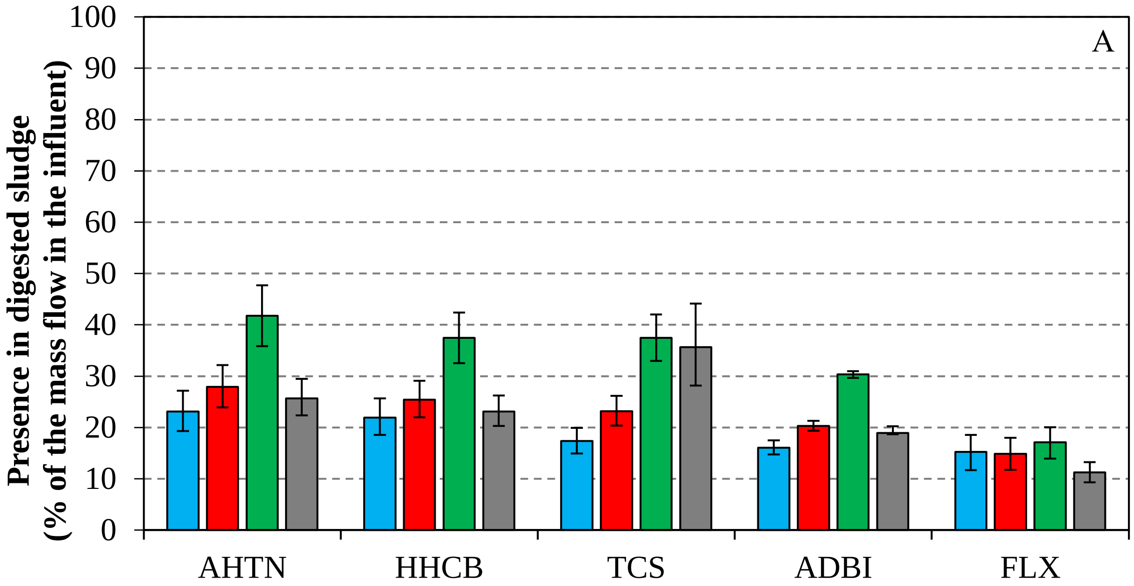

A
High-rate
activated sludge
Settling
tank
Partial nitritation-
anammox
Wastewater
Effluent
7
Thickener
Thermal hydrolysis
Anaerobic digester
Dewatering
Digested
sludge
B
Rotating
belt filter
High-rate activated
sludge reactor
Settling
tank
Partial nitritation-
anammox
Wastewater
Effluent
Thickener
Thermal hydrolysis
Anaerobic digester
Dewatering
Digested
sludge
C
Chemically enhanced
primary treatment
Partial nitritation-
anammox
Wastewater
Effluent
Thickener
Thermal hydrolysis
Anaerobic digester
Dewatering
Digested
sludge
D
Primary
settling
Activated
sludge reactor
Settling
tank
Wastewater
Effluent
Thickener
Anaerobic digester
Dewatering
Digested
sludge

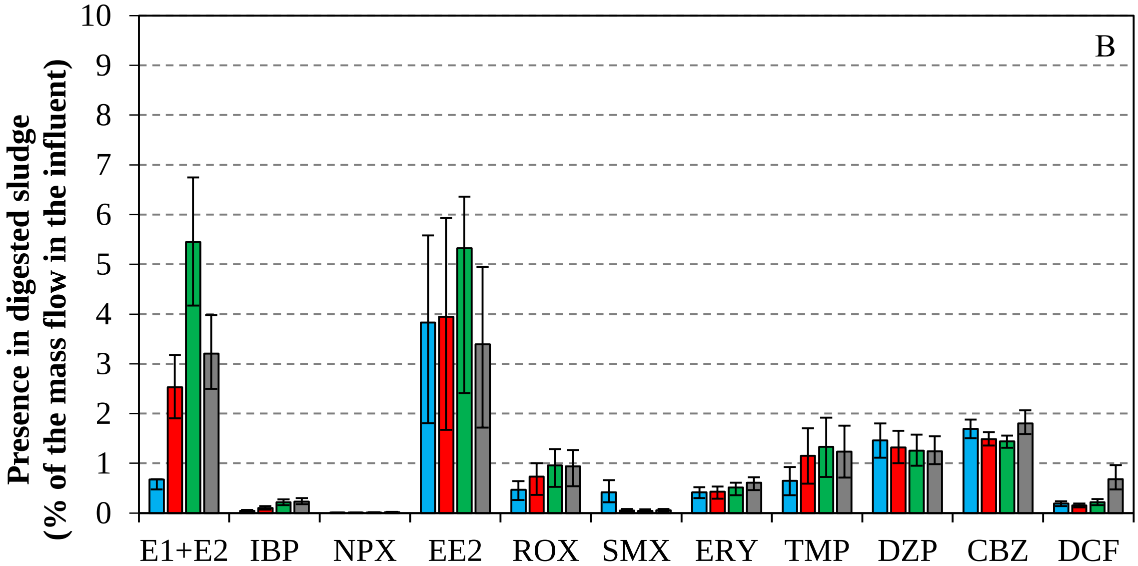
Figure 2.



Removal efficiency from wastewater (%)
100
90
80
70
60
50
40
30
20
10
0
AHTN
HHCB
ADBI
TCS
E1+E2
IBP
NPX
EE2
FLX
ROX
SMX
ERY
TMP
DZP
CBZ
DCF

Figure 3.


A
Presence in digested sludge
(% of the mass flow in the influent)
100
90
80
70
60
50
40
30
20
10
0
AHTN
HHCB
TCS
ADBI
FLX


B
Presence in digested sludge
(% of the mass flow in the influent)
10
9
8
7
6
5
4
3
2
1
0
E1+E2
IBP
NPX
EE2
ROX
SMX
ERY
TMP
DZP
CBZ
DCF