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Combining Thermophilic Aerobic Reactor (TAR) with Mesophilic Anaerobic digestion (MAD) improves the degradation of pharmaceutical compounds

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Abstract: *The removal efficiency of nine pharmaceutical compounds in conventional Mesophilic Anaerobic Digestion (MAD) was compared with Mesophilic Anaerobic Digestion combined with a Thermophilic Aerobic Reactor (MAD-TAR). The semi-industrial scale reactors were fed with primary sludge obtained after decantation of urban wastewater. The highest removal rate of target micropollutants were reported for caffeine (CAF) and sulfamethoxazole (SMX) (>89%) with no significant differences between both processes. MAD-TAR process showed a significant increase of removal efficiency for oxazepam (OXA) (73%), propranolol (PRO) (61%) and OFL (41%) and slightly for diclofenac (DIC) (4%) and 2 hydroxy-ibuprofen (2OH-IBP) (5%). However, ibuprofen (IBP) and carbamazepine (CBZ) were not degraded during both processes.*

Keywords: *pharmaceutical compounds; Thermophilic Aerobic Reactor; Mesophilic Anaerobic Digestion*

Session .. – Anaerobic transformation of micro-pollutants

Introduction

Over the last twenty years, various studies have quantified concentrations and occurrences of pharmaceutical compounds in WWTPs. Depending on the physicochemical properties of the molecules, a significant fraction can adsorb onto the sludge. Anaerobic digestion is the most widely used method for treating (with valorization) sludge produced in waste water treatment plants and the evaluation of the degradability of pharmaceutical molecules during digestion is essential for later recovery of digestate. However, although various studies have investigated the fate of pharmaceutical compounds in the water sector, their behavior during anaerobic digestion has been little studied. In addition, innovation on the process is still needed to improve the degradation of these compounds.

In this work, the degradation of nine compounds (caffeine CAF, ofloxacin OFL, sulfamethoxazole SMX, propranolol PRO, carbamazepine CBZ, oxazepam OXA, diclofenac DIC, ibuprofen IBP and 2 hydroxy-ibuprofen 2OH-IBP) was evaluated. A conventional mesophilic anaerobic digestion (MAD) was compared to a system coupling a thermophilic aerobic reactor (MAD-TAR).

Material and Methods

Biodegradation of the nine molecules was evaluated in an experimental set-up composed of a mesophilic anaerobic digester (MAD) coupled with a thermophilic aerobic digester (TAR). The MAD is a 260 L tank with an agitator and a double jacket connected to the TAR by means of an ALBIN peristaltic recirculation pump (type ALH15), whose flow rate was fixed at 100 L / h. The TAR, with a volume of 40 L, was also equipped with an agitator and a double jacket. The two reactors were heated by two independent cryostats in order to maintain the temperatures suitable for aerobic thermophilic (65 °C) and anaerobic mesophilic (35 °C) processes.

The feed was performed with a primary sludge obtained after decantation of urban wastewater from the Cugnaux WWTP (France) and the duration of feeding, withdrawal and recirculation were adjusted to maintain a total SRT of around 20 days and a residence time in the TAR of 1 day.

Two campaigns were performed during the same period in two consecutive years: one in pure mesophilic anaerobic conditions and one with the hybrid process MAD-TAR.

Quantification of micropollutants was performed by UHPLC-MS-MS following a modified QuEChERS extraction.

Results and Conclusions

Performances of digestion

Although the feeding showed fluctuations, as can be expected when working with real influent, the VSS and COD_t concentrations after digestion remained constant with standard deviation below 4%. Considering comparable operating conditions, higher elimination of solid organic matter was achieved in the hybrid MAD-TAR system (around 76% for VSS) compared to the mesophilic one (around 47%) confirming lab results obtained a few years ago (Dumas et al., 2010). Operational conditions and main results are reported in Table 1.

Table 1 Methanisation performance

	MAD	MAD-TAR
<i>Operational conditions</i>		
OLR (kgCOD/m ³ /day)	1.2	0.9
OLR (kgVSS/m ³ /day)	0.8	0.6
SRT (days)	19.6	22.4
<i>Reactor</i>		
TSS (g/L)	11.7±0.5	4.9±0.6
VSS (g/L)	8.3±0.4	3.4±0.4
COD _t (g/L)	14.1±1.4	5.7±0.8
COD _s (g/L)	0.37±0.06	0.36±0.08
<i>Removal efficiencies (%)</i>		
COD	46.7	72.9
VSS	47.0	75.7

Fate of micropollutants during digestion

The total concentration (µg/L) of the nine selected compounds is illustrated in Figure 1 for both primary sludge and digestate from MAD (Figure 1a) and MADTAR (Figure 1b) processes. All the target molecules were detected and quantified in the primary sludge. Even if some variations have been observed between the two experimental campaigns (-23% in average during the MAD-TAR experiment) the order of magnitude were the same. The highest concentrations of targeted compounds were observed for OFL and CAF (more than 40 µg/L) while medium to low concentrations were reported for SMX, CBZ, PRO, DIC, OXA, IBP and 2OH-IB, around 1.0-10.0 µg/L.

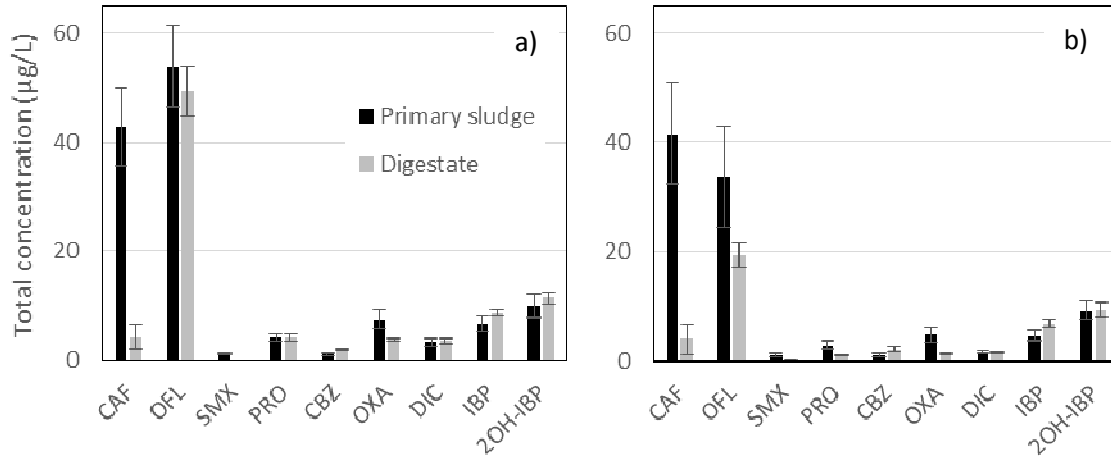


Figure 1 Concentration of targeted compounds in primary sludge and digestate in a) MAD (n=13); b) MAD-TAR (n=13)

The comparison of removal efficiencies in MAD and MAD-TAR are reported in Figure 2. Compounds placed above the dotted line show better removal efficiency by the innovative process (MAD-TAR) compared to conventional process (MAD).

The highest removal rates were obtained for CAF and SMX (>89%) with no significant differences between both processes. The removal of others target compounds was significantly improved by MAD-TAR process. Medium removal ranging between 54% and 75% were observed for OFL and OXA respectively in MAD-TAR compared to only 9% and 49% in MAD. Moreover, while PRO was not removed at all during MAD process, a rate of 69% was achieved in MAD-TAR. DIC and 2OH-IBP present low removal during MADTAR (6-10%) while they were accumulated in MAD. Finally, a negative removal rate was observed for IBP and CBZ in both processes, MAD (-30% and -71%, respectively) and MAD-TAR (-35% and -91%).

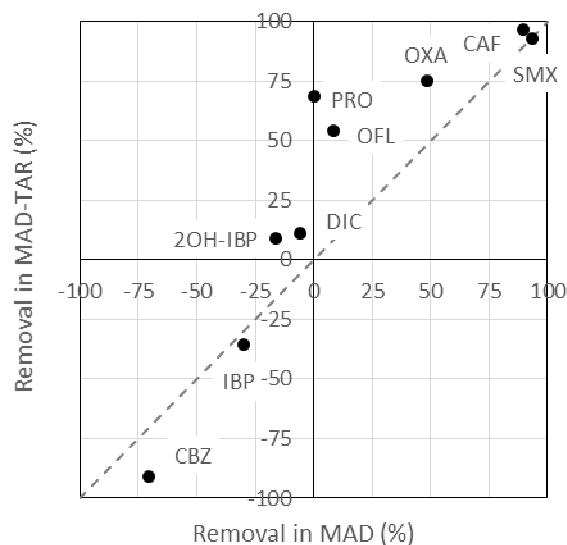


Figure 2 Comparison of compounds removal in conventional vs innovative process

Similar removal efficiencies were reported in the literature for CAF and SMX (Narumiya et al. 2013; Carballa et al. 2007) and OFL (Narumiya et al. 2013), negative removal for CBZ was

also observed by Narumiya et al. 2013. Unlike our results, IBP, PRO and DIC are generally moderate-low eliminated in conventional MAD process (Malmborg and Magnér 2015; Samaras et al. 2014; Martin Ruel et al. 2012; Carballa et al. 2007; Narumiya et al. 2013).

In conclusion, even if non-significant differences were found for CAF and SMX which were well-removed by both processes, the hybrid MAD-TAR process improves significantly the removal efficiency of OXA, PRO and OFL and slightly DIC and 2OH-IBP concomitantly with the improvement of overall organic matter degradation. However, IBP and CBZ even show more negative efficiencies and the mechanisms involved need further investigations.

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