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Unraveling the potential of a combined nitritation-anammox biomass towards the biodegradation of pharmaceutically active compounds

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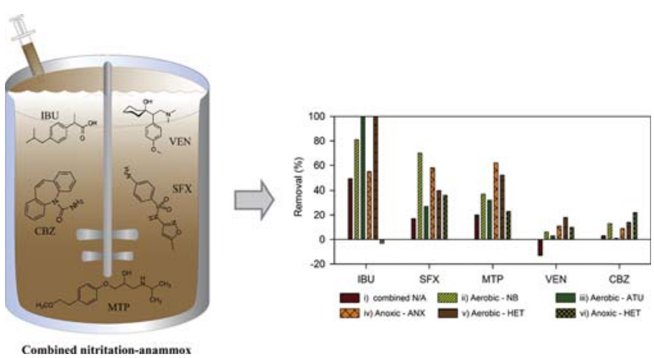
HIGHLIGHTS

- Microbial populations and operational parameters triggered pharmaceuticals removal.
- Ibuprofen was affected by redox conditions and was aerobically removed up to 100%.
- Higher ammonium oxidation rate led to higher removals for three studied compounds.
- Sulfamethoxazole removal decreased significantly when nitrification was inhibited.
- Carbamazepine and venlafaxine displayed a recalcitrant behavior at any condition.

GRAPHICAL ABSTRACT



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ABSTRACT

In the past few years, anaerobic ammonium oxidation-based processes have attracted a lot of attention for their implementation at the mainstream line of wastewater treatment plants, due to the possibility of leading to energy autarky if combined with anaerobic digestion. However, little is known about the potential degradation of micropollutants by the microbial groups responsible of these processes and the few results available are inconclusive. This study aimed to assess the degradation capability of biomass withdrawn from a combined nitritation/anaerobic ammonium oxidation (combined N/A) pilot plant towards five pharmaceutically active compounds (ibuprofen, sulfamethoxazole, metoprolol, venlafaxine and carbamazepine). Batch experiments were performed under different conditions by selectively activating or inhibiting different microbial groups: i) regular combined N/A operation, ii) aerobic (optimal for nitrifying bacteria), iii) aerobic with allylthiourea (an inhibitor of ammonia monooxygenase, enzyme of ammonia oxidizing bacteria), iv) anoxic (optimal for anaerobic ammonium oxidizing bacteria), v) aerobic with acetate (optimal for heterotrophic bacteria) and vi) anoxic with acetate (optimal for heterotrophic denitrifying bacteria). Ibuprofen was the most biodegradable compound being significantly degraded (49–100%) under any condition except heterotrophic denitrification. Sulfamethoxazole, exhibited the highest removal (70%) under optimal conditions for nitrifying bacteria but in the rest of the experiments anoxic conditions were found to be slightly more favorable (up to 58%). For metoprolol the highest performance was obtained under anoxic conditions favoring anammox bacteria (62%). Finally,

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carbamazepine and venlafaxine were hardly removed (≤10% in the majority of cases). Taken together, these results suggest the specificity of different microbial groups that in combination with alternating operational parameters can lead to enhanced removal of some micropollutants.

1. Introduction

The prevalence and continuous input of micropollutants into water bodies is one of the main environmental challenges to be tackled. These compounds exert several toxic biological effects such as estrogenicity, mutagenicity and genotoxicity, but their exact adverse impacts are not yet fully known. Treatment at the source should be favored over centralized end-of-pipe treatment (Joss et al. 2006), but much effort is still needed before this issue can be addressed. Hence, optimization of the current wastewater treatment methods to create a barrier to micropollutants emission remains a high priority task (Luo et al. 2014).

On the other hand, the increasing energy demand due to population growth and high living standards in combination with the need to maintain a reliable effluent quality, arise concerns for many authorities. Therefore, although the main aim of wastewater treatment plants (WWTPs) is to reduce harmful emissions into water bodies, more attention is recently paid to energy efficiency and even broader to overall environmental sustainability (Schaubroeck et al. 2015). Consequently, we need to opt for technologies that could integrate across different aspects, such as addressing micropollutants-related water quality issues at the lowest energy and carbon footprint possible.

Nowadays, the most promising alternative for biological nitrogen removal in WWTPs is the anaerobic ammonium oxidation process since it entails significant advantages over the conventional nitrification-denitrification pathway. It offers up to 60% reduction in aeration requirements and consequently in energy consumption, 100% savings in organic carbon (autotrophic bacteria) and up to 90% reduction in sludge production (Lackner et al. 2014; Laureni et al. 2015; Schaubroeck et al. 2015; van Loosdrecht and Brdjanovic 2014), leading at the same time to a significant reduction in operational costs. Moreover, in combination with anaerobic digestion, the energy balance of wastewater treatment could turn out neutral or even positive (Siegrist et al. 2008). In the past several years, numerous processes under different reactor configurations have been successfully applied under mesophilic conditions at full scale (Lackner et al. 2014) for the treatment of high strength ammonium wastewater (typically the reject water from the anaerobic digester - sidestream operation). However, their potential and reliability in meeting the nitrogen discharge limits during the direct treatment of municipal wastewater (mainstream operation) under psychrophilic conditions have only recently been confirmed (Laureni et al. 2016).

Moreover, many studies have reported higher removals of pharmaceutically active compounds (PhACs) and endocrine disruptors in nitrifying systems and a positive association was demonstrated with the activity of ammonia oxidizing bacteria (AOB) (Fernandez-Fontaina et al. 2012; Kassotaki et al. 2016; Tran et al. 2009; Yi and Harper 2007). Nevertheless, the potential of AOB in combination with anaerobic ammonium oxidizing (anammox) bacteria for the biodegradation of micropollutants has only been investigated in a limited number of studies and up-to-date results are somewhat contradictory. Laureni et al. (2016) stated that the removal of organic micropollutants (including sulfamethoxazole, metoprolol, venlafaxine and carbamazepine) in mainstream combined nitritation/anaerobic ammonium oxidation (combined N/A) systems was comparable to the removal achieved in conventional processes for biological nutrient removal. On the contrary, >80% removal was reported in one stage sidestream combined N/A process for estrone, estradiol, ethinylestradiol, naproxen, ibuprofen, bisphenol A and celestolide (Alvarino et al. 2015). Finally, de Graaff et al. (2011) studied the removal of micropollutants in a two stage combined N/A and reported that the contribution of the anammox reactor was substantial only in the case of ibuprofen, whereas some of the compounds (i.e. propranolol, carbamazepine and cetirizine) exhibited no removal. These results highlight the necessity for more in depth exploration on the potential of the combined N/A process towards the removal of micropollutants.

In the present work, the degradation capability of the microbial biomass collected from a combined N/A pilot plant towards the simultaneous removal of ammonia and five PhACs was studied. To investigate if degradation of the selected compounds will occur and to unravel which microbial groups are mainly involved in this activity, batch experiments were performed under different conditions. Moreover, the formation of several major transformation products (TPs) was investigated. To the best of our knowledge, this is the first time that the different contributions of the microbial groups thriving in a combined N/A bioreactor towards the degradation of PhACs and the consequent formation of TPs is being discussed.

2. Materials and methods

2.1. Pilot-scale reactor operation

Biomass from a 400 L combined N/A pilot-scale reactor of both attached (carrier: FLUOPUR® synthetic porous fleece material, WABAG Water Technology Ltd., Switzerland; 40% fill ratio) and suspended biomass treating digester supernatant was used for the experiments. The combined N/A reactor consisted of a stirrer, a feed pump, a decanter, and an aeration unit and the process - that was automatically controlled - comprised of different phases as described elsewhere (Joss et al. 2011).

2.2. Activity tests

Preliminary assays were performed in order to evaluate the activity of the bacterial communities present in the combined N/A reactor. The maximum activities of the three main autotrophic groups of microorganisms (Anammox, AOB and nitrite oxidizing bacteria-NOB) as well as of the heterotrophic fraction present in the biomass were investigated prior to the execution of the main batch experiments. The maximum anammox activity is defined as the nitrogen removal rate (sum of NH_4^+ and NO_2^-) in the absence of dissolved oxygen (DO) and under non-limiting concentrations of NH_4^+ and NO_2^-. The maximum AOB and NOB activities (under non-limiting DO concentration) are defined as the NH_4^+ removal rate through oxidation by AOB under non-limiting concentration of NH_4^+ and as the NO_3^- production rate through oxidation of NO_2^- by NOB under non-limiting concentration of NO_2^-, respectively. Finally, in order to evaluate the heterotrophic denitrifying activity, the NO_3^- removal rate in the absence of DO and under non-limiting concentrations of NO_3^- and acetate was studied.

For each activity test 10 L of biomass were withdrawn from the pilot-scale 400 L reactor and were transferred to a 12 L reactor that was controlled by a programmable logical controller (PLC) equipped with online sensors (as described in Section 2.3). The three activity assays had different duration and were performed under different initial concentrations of NH_4^+ and NO_2^- taking into account the difference between the yields of the microbial groups as well as the inhibitory effect of NO_2^- to anammox bacteria over specific concentrations as demonstrated in Lackner et al. (2014). NH_4^+ and NO_2^- were supplied as NH_4CL and $NaNO_2$. In the case of the anammox activity test an initial concentration of ≥80 mg N/L for both NH_4^+ and NO_2^- was achieved in the system. For