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Fate of pharmaceuticals in full-scale source separated sanitation system

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ABSTRACT

Removal of 14 pharmaceuticals and 3 of their transformation products was studied in a full-scale source separated sanitation system with separate collection and treatment of black water and grey water. Black water is treated in an up-flow anaerobic sludge blanket (UASB) reactor followed by oxygen-limited autotrophic nitrification–denitrification in a rotating biological contactor and struvite precipitation. Grey water is treated in an aerobic activated sludge process. Concentration of 10 pharmaceuticals and 2 transformation products in black water ranged between low μg/l to low mg/l. Additionally, 5 pharmaceuticals were also present in grey water in low μg/l range. Pharmaceutical influent loads were distributed over two streams, i.e. diclofenac was present for 70% in grey water, while the other compounds were predominantly associated to black water. Removal in the UASB reactor fed with black water exceeded 70% for 9 pharmaceuticals out of the 12 detected, with only two pharmaceuticals removed by sorption to sludge. Ibuprofen and the transformation product of naproxen, desmethylnaproxen, were removed in the rotating biological contactor. In contrast, only paracetamol removal exceeded 90% in the grey water treatment system while removal of other 7 pharmaceuticals was below 40% or even negative. The efficiency of pharmaceutical removal in the source separated sanitation system was compared with removal in the conventional sewage treatment plants. Furthermore, effluent concentrations of black water and grey water treatment systems were compared with predicted no-effect concentrations to assess toxicity of the effluent. Concentrations of diclofenac, ibuprofen and oxazepam in both effluents were higher than predicted no-effect concentrations, indicating the necessity of post-treatment. Ciprofloxacin, metoprolol and propranolol were found in UASB sludge in μg/g range, while pharmaceutical concentrations in struvite did not exceed the detection limits.

1. Introduction

Conventional sanitation systems developed throughout the 20th century use large amount of water in order to transport waste from our houses to the treatment sites. The drawbacks of such systems are high drinking water consumption, low energy efficiency and high construction costs for sewage systems (Verstraete and Vlaeminck, 2011). Moreover, dilution of organic matter and nutrients present in wastewater hinders possibility for their recovery and reuse (Zeeman and Lettinga, 1999). Concentration of organic matter and nutrients in a relatively small volume is achieved by separate collection of toilet wastewater, generally indicated as black water (Otterpohl et al., 1997). Recovery of organic matter and nutrients is possible by application of anaerobic treatment for biogas production from the organic matter and struvite precipitation for fertilizer production (Tervahauta et al., 2013). UASB sludge can also be used as organic fertilizer (Tervahauta et al., 2014a). The rest of the domestic wastewater, generally indicated as grey water, is considered as a relatively clean wastewater stream which can be reused after appropriate treatment (Ghunmi et al., 2011).

Reuse of treated wastewater and its components is possible only when no environmental or health risks are associated to such applications. Apart from being a valuable resource, wastewater contains possible threats for environment and human health, such as pathogenic organisms, heavy metals and persistent organic pollutants (Winker et al., 2009; Tervahauta et al., 2014b).

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Pharmaceuticals are the main group of persistent organic pollutants present in black water (de Graaff et al., 2011). They are excreted from the human body either as parent compounds or their metabolites and end up in the wastewater streams (Snyder et al., 2003). Their concentrations in discharged wastewaters are not regulated by the existing effluent discharge requirements (Sanderson, 2011). However, a number of these compounds exert negative effects on the environment and human health, including feminization of male organisms, growth inhibition, carcinogenicity and bioaccumulation (Halling-Sørensen et al., 1998). Pharmaceuticals are present in conventional wastewater in ng/l to μg/l range and a number of them are not removed in conventional activated sludge treatment systems (Onesios et al., 2009; Verlicchi et al., 2012; Joss et al., 2005). By separate collection of black water pharmaceuticals are concentrated in a relatively small stream that facilitates their removal (Winker et al., 2009; de Graaff et al., 2011; de Mes et al., 2008; Larsen et al., 2009).

de Graaff et al. (2011) studied pharmaceutical removal in a lab-scale black water treatment system, consisting of an up-flow anaerobic sludge blanket (UASB) reactor and a two reactor partial nitritation–anammox process. However, these authors measured concentrations of pharmaceuticals in liquid streams only, disregarding pharmaceutical outflows through UASB sludge, which may be an important pathway for pharmaceuticals entering the environment. Moreover, the post-treatment of black water effluent possibilities were not studied. Thus, more extended research of pharmaceutical removal in the source separated sanitation (SSS) systems is required.

This paper is focused on removal of widely used pharmaceuticals belonging to different therapeutic groups and their transformation products in a full-scale black water treatment system consisting of the UASB reactor, oxygen-limited autotrophic nitrification–denitrification (OLAND) process and struvite precipitation reactor. The aim of the paper is to provide insights on removal and conversion of pharmaceuticals and their transformation products, their incorporation to UASB sludge and struvite, and possibility for the effluent post-treatment in the existing grey water treatment system. Additionally, comparison with the removal in the conventional sewage treatment plants (STPs) is provided and potential ecotoxicity of the effluents is discussed.

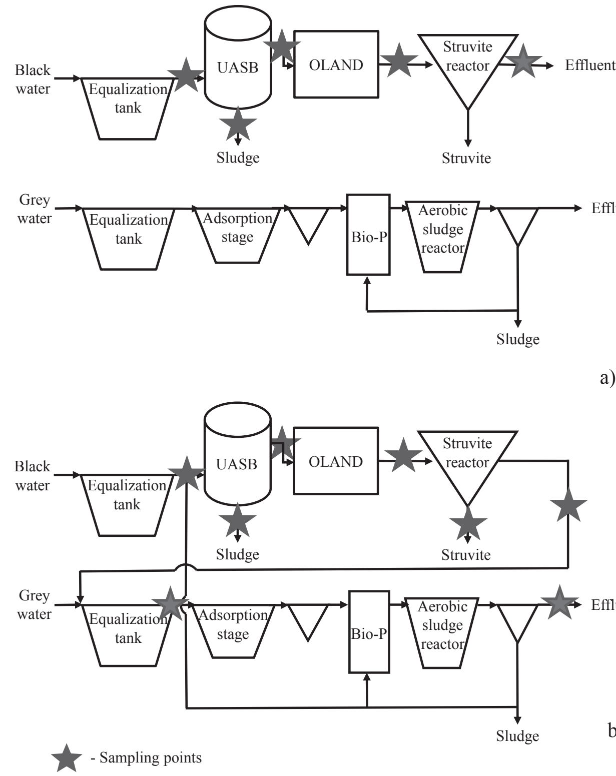

Fig. 1. Source Separated Sanitation system at Noorderhoek. a) is the system with separated grey water and black water treatment; b) is the system with combined black water and grey water treatment.

2. Materials and methods

2.1. Treatment system and sampling

The SSS full-scale demonstration site Noorderhoek (Sneek, the Netherlands) includes separate collection and transport of black and grey water, installed in 62 apartments, and a treatment facility for these separate streams (Fig. 1). The population served by the SSS system consists primarily of elderly people with high use of pharmaceuticals. Black water consists of toilet wastewater and shredded fruit and vegetable waste from the kitchen grinders. Black water is collected by vacuum pumps. The flow is equalized in an underground stirred tank with a hydraulic retention time (HRT) of 0.5 d. Black water is anaerobically digested in a UASB reactor (T = 32 °C, HRT = 35 d). The effluent of the UASB reactor is treated with OLAND in a rotating biological contactor (biorotor). The biorotor is followed by magnesium ammonium phosphate (struvite) precipitation (STOWA, 2014).

The grey water is collected separately from black water and the flow is equalized in an underground stirred tank (HRT = 24 h). Grey water treatment system was originally designed as an adsorption/biooxidation (AB) system, consisting of the high-loaded aerated adsorption stage and a low-loaded biooxidation stage. During the sampling campaign the adsorption stage was not functioning, serving as an extension of the equalization tank with an HRT of 22 h. The biooxidation stage was thus serving as an aerobic activated sludge treatment system (HRT = 10 h), preceded by an anaerobic compartment for enhanced biological phosphorus removal and followed by a settler.

The effluent of the black water treatment system has been added to the equalization tank of the grey water treatment system from 16th May 2013 in order to use it as a post-treatment step. The sludge from the activated sludge grey water treatment system has been added to the UASB reactor from 11th July 2013 in order to increase energy efficiency of the concept.

The sampling was performed in two periods (5th March–30th May 2013 and 26th August–2nd October 2014), thus, on separate and combined wastewater streams. The average black water flow was 1.1 m^3/d during both sampling periods. The average grey water flow was 5.0 m^3/d during the first sampling period and 5.7 m^3/d during the second sampling period.

Grab samples for pharmaceuticals analysis in the black water treatment system were taken from the sampling taps located after the equalization tank, the UASB reactor, the biorotor and the struvite precipitation unit. UASB sludge was sampled from the lowest tap of the UASB reactor (h = 1 m). Struvite was sampled directly from the precipitation unit only during the second sampling period. Grab samples for pharmaceuticals analysis in the grey water treatment system were taken from the sampling taps located after the equalization tank and after the settler of the activated sludge treatment system. These samples were taken only during the second sampling period to assess pharmaceutical removal in the grey water treatment system after black water addition.

Fifteen samples (V = 30 ml) were taken at each sampling point during each sampling period. All samples were taken randomly at