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Detailed mass flows and removal efficiencies for biocides and antibiotics in Swedish sewage treatment plants

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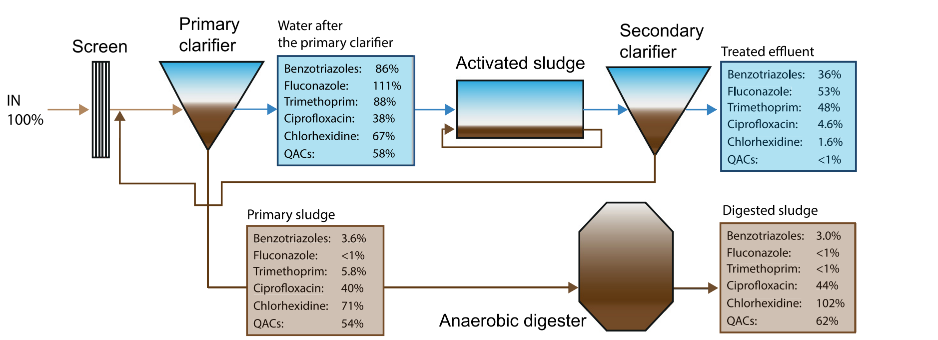
HIGHLIGHTS

- Detailed mass flows for eleven antimicrobials in different sections of three STPs.
- First mass balance reported for chlorhexidine and hexadecylpyridinium chloride.
- No biodegradation of chlorhexidine.
- The biological treatment step was the most important for degradation.

GRAPHICAL ABSTRACT



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ABSTRACT

Antimicrobial compounds, such as biocides and antibiotics, are widely used in society with significant quantities of these chemicals ending up in sewage treatment plants (STPs). In this study, mass flows and removal efficiency in different treatment steps at three Swedish STPs were evaluated for eleven different biocides and antibiotics. Mass flows were calculated at eight different locations (incoming wastewater, water after the first sedimentation step, treated effluent, primary sludge, surplus sludge, digested sludge, dewatered digested sludge and reject water). Samples were collected for a total of nine days over three weeks. The STPs were able to remove 53–>99% of the antimicrobial compounds and 0–64% were biodegraded on average in the three STPs. Quaternary ammonium compounds were removed from the wastewater >99%, partly through biodegradation, but 38–96% remained in the digested sludge. Chlorhexidine was not biodegraded but was efficiently removed from the wastewater to the sludge. The biological treatment step was the most important step for the degradation of the studied compounds, but also removed several compounds through the surplus sludge. Compounds that were inefficiently removed included benzotriazoles, trimethoprim and fluconazole. The study provides mass flows and removal efficiencies for several compounds that have been seldom studied.

1. Introduction

Various forms of micropollutants are constantly entering the aquatic environment because of incomplete removal by sewage treatment plants (STPs) (Kümmerer, 2009; Loos et al., 2013; Luo et al., 2014). Chemicals requiring more attention in wastewater studies are antimicrobial biocides. Biocides have a wide application in society as preservatives, disinfectants and antiseptics (Wieck et al., 2018, 2016). Several of them are high production volume chemicals with large numbers of applications in both the household and industrial sectors. The presence of a number of biocides in STPs has been reported several times (Bollmann et al., 2014; Liu et al., 2017; Östman et al., 2017). Concerns have been raised that the use of these biocides might be linked to the immense

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global antibiotic resistance problem (SCENIHR, 2009). Antibiotic resistance results in >700,000 deaths annually and it is expected to increase massively (O'Neill, 2014). The current resistance problem in human pathogens is mainly driven by the overuse and misuse of antibiotics but biocides may also contribute through co-resistance and cross-resistance (SCENIHR, 2009). STPs have been identified as potential hotspots for resistance development (Berendonk et al., 2015; Berglund et al., 2015; Guo et al., 2017) which makes it important to understand the fate of antimicrobial compounds in the wastewater treatment processes. Several studies exist that have determined the fate of antimicrobials in STPs (Castiglioni et al., 2018; Clara et al., 2007; Göbel et al., 2007; Guerra et al., 2014; Heidler and Halden, 2008; Lindberg et al., 2005, 2006; Liu et al., 2017). Most of these studies have focused predominantly on antibiotics and less on biocides. Furthermore, the majority of these studies have usually only considered the final products of the STPs to measure the overall mass balance and removal efficiency, and have done less monitoring of what is happening at the different treatment steps. In a previous study, we detected several biocides that had been seldom studied in STPs in the past, such as chlorhexidine, hexadecylpyridinium chloride and hexadecyltrimethylammonium bromide (CTAB) (Östman et al., 2017). However, data are missing about the fate of these compounds at different treatment steps in the STPs. The aim of this study was, therefore, to make a detailed mass balance study of a number of antimicrobial compounds within three Swedish sewage treatment plants. Apart from the seldom studied compounds, other quaternary ammonium compounds (QACs) were included as well as some better studied compounds such as trimethoprim, ciprofloxacin and fluconazole. The three STPs were of different sizes and slightly different configurations and were sampled in 2015.

2. Materials and methods

2.1. Selection of compounds

The selections of biocides and antibiotic compounds were based on previous findings in STPs, usage data from Sweden, stability and sales. See (Östman et al., 2017) for a full description of the selection process. In this study, we made a selection of the most common compounds detected in STPs from our previous study, to ensure it was possible to calculate a good mass balance. A list of the selected compounds is shown in Table 1.

2.2. Studied sewage treatment plants

Three STPs of various sizes, different treatment configurations and geographical locations were included in the study. A schematic overview of the treatment steps at each STP and corresponding retention times is shown in Fig. 1. The STPs are described below and additional parameters and exact locations can be found in the supplementary information in Fig. S1, Table S1 and S2, respectively. No changes have been made to the STPs in the study.

2.2.1. BROMMA STP (STOCKHOLM)

Bromma STP is located in Stockholm and serves approximately 350,000 inhabitants (208,000 pe) in the western part of the city (*Environmental report from Stockholm vatten 2015*, 2016). The STP treats the water using mechanical, chemical and biological steps with nitrogen removal. For the first step, $FeSO_4$ is added for flocculation, followed by the use of a 3 mm step screen to remove larger objects and then a sand trap with 3 min retention time. The water is aerated before it enters the primary clarifier where a large portion of the solids are removed through sedimentation in 24 pools. The water is then pumped to the biological treatment step, which consists of active sludge treatment in six pools for 3.6 h followed by 12 secondary clarifiers with a retention time of 4 h. As a final step, the water passes through sand filters before it is released into the Baltic Sea. The primary sludge from the primary clarifiers is pumped to digestion chambers, maintained at a temperature of 34–37 °C, where the sludge remains for a minimum of 15 days. Surplus sludge coming from the activated sludge treatment and secondary clarifiers is dewatered using centrifugation, as well as being digested. The digested sludge is dewatered using centrifugation.

2.2.2. RYA STP (GÖTEBORG)

Rya STP, located in Göteborg, serves 737,000 inhabitants (806,575 pe) and is the largest sewage treatment plant in Sweden with respect to the volume of incoming water (Mattsson, 2016). About 50% of the incoming water comes from stormwater and water leaking into the pipes. As can be seen in Fig. 1, the wastewater is treated mechanically with a coarse screen with 20 mm spacing followed by an aerated sand trap and a 2 mm fine screen, before it reaches the primary clarifiers that have a retention time of 1.7 h. The primary sludge from the clarifier is thickened before it is pumped to the digestion chamber, where it is digested for 20 days at 37 °C. The water from the primary clarifiers is pumped to the activated sludge treatment. If necessary, $FeSO_4$ is used as a flocculation treatment before the active sludge. After the activated sludge treatment (1.5 h retention time), the water goes to the secondary clarifiers for 1.5 h. The majority of the sludge from the secondary clarifiers is recirculated to the active sludge treatment and a small part is pumped back to the primary clarifiers. Approximately half of the water from the secondary clarifier is passed through disc filters (15 μm) and released into the receiving body of water. The other half goes through a nitrogen removal step using nitrifying trickling filters and post-denitrification before it goes to the disc filters and is released into the mouth of the river Göta älv, close to the sea.

2.2.3. ÖN STP (UMEÅ)

Ön STP treats the wastewater from 96,000 people (83,919 pe) in Umeå in northern Sweden (Nordlund, 2016). Due to cold weather, the plant has no treatment step to remove nitrogen but otherwise treats the water mechanically, chemically and biologically (see Fig. 1). The incoming wastewater is passed through screens with 2 mm spacing followed by aerated sand removal (12 min retention time). The chemical treatment step consists of flocculation pools and primary clarifiers with 1.2 h retention time. $FeCl_3$ is used for flocculation treatment. The biological treatment step consists of an active sludge treatment (6 h retention time) after the chemical treatment. Before the water is released into the Ume river, there is another chemical treatment step, with flocculation (using $FeCl_3$) in pools followed by secondary clarifiers with 2.6 h retention time. Part of the sludge from the active sludge treatment is recirculated and the surplus sludge is pumped to a point before the primary clarifiers. The sludge from the secondary clarifiers is also pumped to a point before the primary clarifiers. The primary sludge from the primary clarifiers is a mixed sludge, since it contains contributions from the surplus sludge and sludge from the secondary clarifier. The primary sludge is thickened before it enters the digestion chambers where it is digested for 25–30 days at 37 °C. The digested sludge is then dewatered using centrifugation.

2.3. Sampling and sample handling

The concentration of different micropollutants have been shown to both increase (Bollmann et al., 2014) and decrease (Benotti and Brownawell, 2007) during rainfall. Although the mass balance, in theory, is not affected, the function of the STP can be affected with too much water. To minimize the impact of variations, the STPs were sampled on three consecutive days each week over 3 weeks ($n = 9$) in January–March 2015 (see Tables S3–S5 for sampling dates). Incoming sewage water, treated effluent and water after the primary clarifier were sampled as 24 h composite samples using the automatic samplers at the sewage treatment plants. Rya STP used flow proportional samplers and Bromma and Ön STPs used volume proportional samplers according to the definition given by (Ort et al., 2010). Sludge was taken as