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Occurrence, identification and removal of microplastic particles and fibers in conventional activated sludge process and advanced MBR technology

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ABSTRACT

Wastewater treatment plants (WWTPs) are acting as routes of microplastics (MPs) to the environment, hence the urgent need to examine MPs in wastewaters and different types of sludge through sampling campaigns covering extended periods of time. In this study, the efficiency of a municipal WWTP to remove MPs from wastewater was studied by collecting wastewater and sludge samples once in every two weeks during a 3-month sampling campaign. The WWTP was operated based on the conventional activated sludge (CAS) process and a pilot-scale membrane bioreactor (MBR). The microplastic particles and fibers from both water and sludge samples were identified by using an optical microscope, Fourier Transform Infrared (FTIR) microscope and Raman microscope. Overall, the retention capacity of microplastics in the studied WWTP was found to be 98.3%. Most of the MP fraction was removed before the activated sludge process. The efficiency of an advanced membrane bioreactor (MBR) technology was also examined. The main related finding is that MBR permeate contained 0.4 MP/L in comparison with the final effluent of the CAS process (1.0 MP/L). According to this study, both microplastic fibers and particles are discharged from the WWTP to the aquatic environment.

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1. Introduction

Investigations related to the occurrence and fate of microplastics (MPs) in aquatic environment have been accelerating worldwide especially during the last decade (Andrady, 2011; Hidalgo-Ruz et al., 2012), considering the increase in plastic production (PlasticsEurope, 2016) and consequently the risk of small plastic particles contaminating our environment. Microplastics may be discharged to the environment from plastic industries e.g. via leakages and transportation accidents. Other sources include the tear and wear of plastic items, the use of personal care products (Fendall and Sewell, 2009) and washing of synthetic textiles (Browne et al., 2011).

One important reason for the urgent need to promote research studies on microplastics is their partly unknown potential to cause ecotoxicological issues in the environment (Andrady, 2011; Hidalgo-Ruz et al., 2012). In this context, microplastics may act as vectors for harmful additives and contaminants (Teuten et al., 2009) and they have the potential to be transferred within the planktonic food web (Setälä et al., 2014), which might affect the environmental fate of numerous toxic substances.

Research efforts on the behavior and impact of microplastics in aquatic environment seem to be versatile, which makes comparative studies challenging (Hidalgo-Ruz et al., 2012; Underwood et al., 2017). For instance, the classification of microplastics by size differs between studies, but usually the upper size limit is set to 5 mm (Arthur et al., 2009). The lower size limit of studied microplastics often follows the mesh size of Neuston net, starting from 300 μm (Arthur et al., 2009; Hidalgo-Ruz et al., 2012). As the increasing amount of microplastics found in the environment have been noticed to belong to even smaller size range (Klein et al., 2015; Ziajahromi et al., 2017) the lower size limit was decreased below 50 μm (Carr et al., 2016; Talvitie et al., 2015, 2017a) and even to the range of nanoplastics (da Costa et al., 2016).

Microplastics have also been isolated from environmental samples by using variable approaches (Hidalgo-Ruz et al., 2012) and

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Nomenclature

CAS	conventional activated sludge
dw	dry weight
FTIR	Fourier Transform infrared
MBR	membrane bioreactor
MPF	microplastic fiber
MPP	microplastic particle
MP	microplastic
PA	polyamide
PE	polyethylene
PES	polyester
PET	polyethylene terephthalate
PP	polypropylene
SS	suspended solids
SVI	sludge volume index
WAS	waste activated sludge
WPO	Wet peroxide oxidation
WWTP	wastewater treatment plant

then identified either visually or using more reliable identification analyses (Andrady, 2011; Hidalgo-Ruz et al., 2012). Some results are based on a visual identification only (Mason et al., 2016; Michielssen et al., 2016; Talvitie et al., 2015), some have identified all particles and fibers with Fourier transform infrared spectroscopy (FTIR) or Raman spectroscopy (Browne et al., 2011; Murphy et al., 2016; Talvitie et al., 2017a; Ziajahromi et al., 2017) and others have relied on a combined methodology of identification by defining possible microplastics visually and then analyzing only part of them (Carr et al., 2016; Magnusson and Norén, 2014; Mintenig et al., 2017).

Even though the abundance of microplastics in the marine environment has been studied quite extensively for the last few decades (Hidalgo-Ruz et al., 2012), the special case of wastewater treatment plants (WWTP), as a route of microplastics, have started to gain more attention only during recent years (Carr et al., 2016; Dris et al., 2015; Magnusson and Norén, 2014; Mahon et al., 2017; Mason et al., 2016; Michielssen et al., 2016; Mintenig et al., 2017; Murphy et al., 2016; Talvitie et al., 2015, 2017b). Still, only few research investigations have focused on the removal efficiency of microplastics in WWTPs and studied microplastics in other WWTP matrices than final effluent (Carr et al., 2016; Mintenig et al., 2017; Murphy et al., 2016; Talvitie et al., 2017a). First studies have shown that microplastics are effectively removed from wastewater and most of the particles end up in the sludge (Magnusson and Norén, 2014; Michielssen et al., 2016; Talvitie et al., 2015, 2017a). Even though the removal efficiency of microplastics in WWTPs is found to be around 99% (Carr et al., 2016; Magnusson and Norén, 2014; Murphy et al., 2016), microplastics are continuously being discharged from WWTPs to recipient waters. On the other hand, if most of the microplastics are transferred to WWTP sludge, which can be further used in green construction and as a fertilizer in fields, sludge might act as a significant route of microplastics to the environment (Talvitie et al., 2017a; Zubris and Richards, 2005).

Furthermore, most of the researchers have based their results on wastewater or sludge samples collected only for a short period of time. Murphy et al. (2016) collected water and sludge samples from different stages of a large secondary WWTP within couple of days. Mintenig et al. (2017) studied effluents and sludge from several WWTPs in Germany and each sample was collected during one day. Carr et al. (2016) studied wastewater and sludge samples from different stages of several WWTPs in the United States and samples were collected for maximum 12 days during less than one and a half months. Talvitie et al. (2017a) collected sludge and water samples for microlitter studies either once or thrice during one week from different stages of WWTP in Finland. In this regard, our research group and many scientists in the field are emphasizing upon the necessity to analyze microplastic pollution from WWTPs for a longer time period to reveal the temporal variation in microplastic concentrations (Magnusson and Norén, 2014; Murphy et al., 2016; Ziajahromi et al., 2017).

In this context, the aim of this study was to investigate the efficiency of a WWTP based on conventional activated sludge (CAS) process to remove microplastics from wastewater. The abundance of microplastic particles and fibers was monitored in 1) the pretreated influent after primary screening, throughout the different stages of the conventional treatment i.e. 2) effluents from primary clarification and 3) activated sludge process, as well as in 4) the digested sludge, 5) final effluent and 6) the recipient water body close to the outlet of treated wastewater. Furthermore, the efficiency of a pilot-scale membrane bioreactor (MBR) incorporated with the WWTP was examined by sampling permeate and sludge with the purpose of comparing the removal efficiencies of all the treatment processes operating in the plant. Overall, this study was carried out for 3 months with sampling campaigns every two weeks. To our knowledge, this is the first study to monitor the occurrence of microplastic particles and fibers in WWTPs over several months and thus to provide reliable assessment about the efficiency of WWTP to remove microplastics.

2. Materials and methods

2.1. Description of WWTP and MBR pilot plant

In this study, wastewater and sludge samples were collected from the municipal WWTP (Kenkäveronniemi WWTP) located near the city center of Mikkeli, Finland. The WWTP treats approximately 10 000 m^3 of municipal wastewater daily and its treatment processes include screening with step screen and 6 mm sieve, grit separation, primary clarification, biological treatment with activated sludge, final sedimentation and disinfection. During the sampling campaign sludge volume index (SVI) of the activated sludge was approximately 62 mL/g, total suspended solids (SS) for effluent was approximately 10.3 mg/L and SS removal efficiency 98.6%, according to the WWTP's monitoring records. Effluent waters from WWTP are discharged into the nearby Launialanselkä basin (Lake Saimaa) via a 110-meter effluent pipe.

A pilot-scale membrane bioreactor (ARTAS Ltd., Turkey) operating in Mikkeli WWTP since 2014, is able to treat around 3 m^3 of wastewater per day. The MBR pilot plant consists of an anaerobic tank, an aerobic tank and a membrane filtration tank with a submerged MBR unit. The pore size of the submerged flat-sheet membrane units is 0.4 μm (KUBOTA Corporation, Japan). Detailed information about the setup, operating conditions and water treatment efficiency of MBR pilot is available in Gurung et al. (2016, 2017).

2.2. Sample collection

Samples were collected from Kenkäveronniemi WWTP every two weeks between 10th of October 2016 and 2nd of January 2017. Samples were collected on Mondays between 8 and 12 a.m. Preliminary sampling tests were carried out before the actual sampling campaign in order to optimize the sampling methods and mitigate potential sample contamination during the in-site operations, samples transfer and laboratory analyses.

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