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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.

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.

Water samples with a volume of 4.0–30.0 L were collected from influent water (after 6 mm screen, in the beginning of the grit separation basin), after the primary clarification and after the disinfection (Fig. S1). Permeate from MBR pilot plant and lake water from the nearby shore (about 100 m from the effluent outlet) were also studied. It has to be noted that in the present investigation, focusing the efficiency of the WWTP to remove MPs, the nearby lake water was sampled only to estimate the amount of MPs in close proximity to the discharge location of treated wastewater. Hence, the reported concentrations are neither representing the average MPs concentration in the recipient lake, nor being correlated with the one discharged from the WWTP.

Samples were collected with a 10-L stainless steel bucket attached to a metal wire and poured to a cascade of two test sieves with mesh sizes of 0.25 and 5.0 mm. Materials retained on the 0.25 mm sieve were rinsed with distilled water to previously weighed glass beakers with the help of a metal funnel. Equipment was rinsed three times with distilled water to ensure that all particles were transferred properly. Distilled water was stored in a 10-L polyethylene container and in polyethylene squirt bottles. All samples were collected with same set of equipment and sampling was started from points with presumably smallest MP concentrations to decrease the possibility of cross-contamination.

Sludge samples (150–200 mL) were collected with a 10-L stainless steel bucket or a 0.25-L stainless steel cup from activated sludge after the aeration basin, MBR sludge and digested sludge. Samples were immediately poured to glass flasks with the help of a metal funnel. All vessels were sealed with aluminum foil and rubber bands, transferred to the laboratory and stored at 4 °C and dark until further treatments.

To assess possible contamination, field blank samples were treated exactly in the same manner as the other samples by using distilled water instead of wastewater and sludge. Furthermore, for water blank samples, the cascade of sieves were rinsed with distilled water and the 0.25 mm test sieve was rinsed to previously weighed glass beakers. For sludge blank samples, a small amount of distilled water was poured to glass flasks with the help of a metal funnel. Field blank samples were subjected to the before-mentioned treatments.

2.3. Sample pretreatment

2.3.1. Water and wastewater

Water samples were treated according to the method proposed by Masura et al. (2015), with some adjustments. No density separation was performed in order to avoid the loss of any high-density microplastics (Shim et al., 2016), such as polyester with density of 1.24–2.3 g/cm^3 (Hidalgo-Ruz et al., 2012). Furthermore, as cellulose fibers (density 1.5 g/cm^3) are one of the most abundant non-plastic groups in WWTP samples, density separation would not have been suitable for separating them from microplastics due to the overlapping densities of these polymers.

First, the sieved samples were placed in 75 °C drying oven for at least 40 h until totally dry. Then the dried samples (0.15 ± 0.06 g dw) were weighed to determine the required amount of chemicals for the subsequent wet peroxide oxidation (WPO). The oxidation process was carried out by adding 20 mL of aqueous 0.05 M Fe(II) solution and 20 mL of 30% hydrogen peroxide (H_2O_2) to the dried samples. Mixture was left to settle down for 5 min in room temperature, and then the beaker was covered with a watch glass, the mixture was agitated and heated up to 75 °C using hotplate with magnetic stirrer. Depending on the organic matter content in water, the oxidative reaction frequently overflowed the beaker. In this case, small amount of distilled water was added to cool down the reaction. For the influent samples, 40 mL of H_2O_2 were initially added, then another 20 mL of H_2O_2 were supplemented after 20 min in order to promote the degradation of the organic matter. The WPO reaction was conducted for additional 30 min after additions of H_2O_2 or distilled water.

Samples were let to cool down and vacuum filtrated through gridded membrane filters (Sartorius, cellulose nitrate filter, porosity 0.8 μm) with glass fiber filters (VWR, Grade 696, porosity 1.5 μm) at the bottom for mechanical support. Filters were placed in petri dishes and let dry for at least 24 h at room temperature, while being loosely covered with aluminum foil to avoid potential contamination.

Suitability of WPO treatment for MPs was tested with commercial PET and PS beads before sampling campaign and no visible deformations were noticed in treated microplastics.

Due to the large amount of cellulose fibers probably originating mainly from decomposed toilet papers caught in influent samples, samples of 0.25 g dw (corresponding to 0.8–3.0 L) were treated with WPO (Masura et al., 2015) along with cellulase enzymes using the method proposed by Champagne and Li (2009). These samples were transferred with a metallic spatula into glass beakers, treated with WPO, sieved using a 38 μm mesh size sieve and rinsed to glass flasks with a small amount of distilled water. Samples were sealed with foil and stored in the absence of light at 4 °C. Enzymatic degradation was proceeded by adding 250 mg of cellulase (*Aspergillus niger*, 0.8 activity units per mg) to 250 mg dw of influent sample, corresponding to an enzyme loading of 800 activity units/g substrate. Next, 12.5 mL of sodium citrate buffer (pH 4.8) was added to the samples, which were then placed in a temperature-controlled shaker at 40 °C with 160 rpm for 24 h. With varying weights of the influent samples, the amount of enzymes and buffer solution were adjusted to correspond the before-mentioned ratios.

2.3.2. Sludge

Sludge samples were treated with the method described by Murphy et al. (2016). Sludge samples were mixed thoroughly with glass stirring rod, after which subsamples (0.1 g dw) were poured to glass petri dishes and dried at 45 °C for 18 h with pierced foil cover. The dry weights of samples were estimated on a basis of water contents in each sludge type. The target wet weights of subsamples for activated sludge, digested sludge and MBR sludge were 20 g, 3 g and 10 g, respectively.

2.4. Examination and identification of microplastics

2.4.1. Optical microscopy

All samples were visually examined under digital optical microscope (Zeiss, SteREO discovery.V8 with Axiocam 503 color) with maximum magnification of 50×. All possible microplastics and all particles and fibers without a clear organic origin (seeds, cellulose fibers from toilet paper) were collected with tweezers, placed on a paper in another petri dish, and analyzed via successive magnifications. All collected particles and fibers, still including both plastic and non-plastic material, were subjected to a second round of visual identification in which all recovered particles and fibers were photographed and measured for the largest dimension using the built-in ZEN 2.3 software. Particles and fibers, approx. 5200 altogether, were classified visually in 54 groups according to the helicity of the fibers and shapes of cross-section e.g. round, oval or flat, and end of fibers such as clear cut, tapered or frayed, as well as hardness and luster of fragments (Norén, 2007). Representative samples of each group were collected on a microscope slide and further examined using an optical microscope (Zeiss, AxioScope.A1) with a magnification up to 1000× and later with FTIR and/or Raman.

A small amount of distilled water was added to influent samples

to avoid the static electricity of dry cellulose fibers during examination.

For the dried sludge samples, small amount of distilled water was added to break up the sludge material, and then each sample was cautiously examined using tweezers, three times altogether (Murphy et al., 2016). As a slight improvement to this method, the first two examinations were conducted with white background and the third examination with black background (i.e. under the glass dish). Such approach helped recovering microplastics with different colors more easily.

2.4.2. FTIR microscopy and Raman spectroscopy

Representative samples of most of the above mentioned groups (see section 2.4.1) were analyzed with Fourier Transform Infrared (FTIR) microscopy and/or micro Raman spectroscopy, in order to confirm their plastic- or non-plastic nature. The same approach was also discussed by Shim et al. (2017) to generate more reliable results than a single analytical method. For minimizing the chance for false-negative microplastics (Lenz et al., 2015), representatives of all classified particles and fibers (e.g. cellulose) were confirmed with spectroscopic methods. Only particles, which were visually identified to clearly consist of organic or other non-plastic compounds, such as alga, sand and glass, were excluded from the FTIR and Raman analyses. Thus on average 2 to 3 particles or fibers were examined with FTIR microscope and micro-Raman for each group, corresponding to 1.3% and 1.4% of all collected particles and fibers.

Representative samples of most abundant groups (i.e. supposed plastic and other materials) were first analyzed with FTIR microscope (Spotlight 200i FT-IR microscope system equipped with Spectrum Two, Perkin Elmer) in the reflectance mode. 24 scans were taken to produce the spectra with wavelengths between 600 and 4000 cm^{-1} and with the spectral resolution of 4 cm^{-1}. Spectra were compared to the spectra libraries supplied by PerkinElmer.

For the micro-Raman spectroscopy (Horiba Jobin Yvon, Labram HR), the analyses were carried out using the green laser (514.53 nm) and LabSpec 5 software, with spectra covering a wavelength range between 200 and 3000 cm^{-1}. Reference libraries of FTIR and Raman spectra for different polymers and natural fibers were collected from various sources (Crawford and Quinn, 2017; Frère et al., 2016; Zhang et al., 2017).

2.5. Contamination control

In the present work, a serious consideration was given to the risk of contamination (Hidalgo-Ruz et al., 2012; Murphy et al., 2016). Thus, nitrile gloves and 100% cotton clothes were worn as topmost clothes when collecting or treating the samples. Only glass and metal dishes were used, plastic caps were replaced with aluminum foil and all vessels were covered whenever possible. Filters and petri dishes were always examined under the optical microscope before the use in order to remove all visible particles and fibers. Laboratory benches were wiped with cellulose tissue and distilled water thrice before samples were treated. One field blank sample for both water and sludge were collected during each sampling to estimate the amount of contamination during sampling and sample preparation. All used PE-containers and bottles were examined not to act as a source of contamination by filtering stored distilled water once during the sampling campaign with three replicates for each container.

3. Results and discussion

3.1. Contamination

Despite the efforts to decrease contamination, on average, 4.4 (±1.4) and 0.1 (±0.1) microplastics were counted from field blank samples for water and sludge, respectively. As blank samples were not conducted with a known volume of distilled water, contamination is presented as number of MPs per sample. Highest level of contamination was measured for the first sampling event. Some evidence of cross-contamination, such as shells of planktonic organisms, were found from field blank samples collected after lake water sampling. However, the origin of the microplastics contamination could not be assessed without separate control samples for each preparation step. Challenges related to contamination and sample pretreatment are discussed more detailed in supporting information (SI).

3.2. Determination of polymer types

Different polymers were detected after the recovery of microplastic fibers (MPFs) and particles (MPPs) from wastewater, sludge and lake water samples (as illustrated in Fig. 1 and Table 1). The list includes polyester (PES), polyethylene (PE), polyamide (PA) and polypropylene (PP). These polymers were frequently identified and confirmed with FTIR microscope and micro-Raman spectroscopy. Microscopic images are shown in Fig. 2.

The proportion of different polymers varied only little between different sampling dates, when microplastics from all sampling points were considered. During the whole sampling campaign, polyester fibers accounted for 96.3% of the MPFs being equivalent to 79.1% of the total amount of MPs collected from all of the samples. Most of the rest MPFs were made of polyamide (3.1% of all MPFs). Microplastic fibers are usually described to be equally thick with three-dimensional bending (Norén, 2007), which differs them from cellulose-based fibers with ribbon like appearance (Dyachenko et al., 2017; Murphy et al., 2016). In this study also polyester fibers with flat, cotton like appearance, were found (Fig. 2D).

For the MPPs, polyethylene was the most abundant polymer that accounted for 63.9% of MPPs, i.e. 11.4% of the total MPs. Rest of the collected MPPs (36.1%) were identified to consist of polyethylene, polyester and polypropylene.

The results of this study are in accordance with previous studies conducted in WWTPs. Ziajahromi et al. (2017) reported PET fibers and PE particles as most abundant microplastics in effluents of different WWTPs. Polyester was detected most frequently also in the final effluents of a Scottish (28%) (Murphy et al., 2016) and an Australian WWTP (67%) (Browne et al., 2011).

Dull and unevenly shaped polyethylene fragments were present in treated wastewater effluent, similarly with the observations of Talvitie et al. (2017a). The same team indicated that these fragments were most probably originated from cleansing scrubs. Interestingly, in the current study this kind of PE fragments were mostly collected from final effluent and even lake samples, but less frequently from influent or sludge samples. The reason for not finding PE fragments from influent flow could be related to the diurnal variation in the wastewater flow together with the hydraulic retention time in WWTP processes. Such observation suggests that at least part of these cosmetics-originated fragments are passing through the treatment processes in the studied WWTP. Murphy et al. (2016) did not found any PE microbeads in the effluent of secondary WWTP in Scotland. Rather, it was noticed that PE beads were efficiently removed together with grease from the surface of the wastewater. Based on these few studies, the fate of PE fragments tend to vary with the various removal processes in WWTPs.

Along with microplastics, and despite the rigorous oxidation treatment, substantial amounts of organic particles and fibers were abundant in both sludge and WPO-treated wastewater samples. Thus, among this group of non-plastic compounds, many colorful

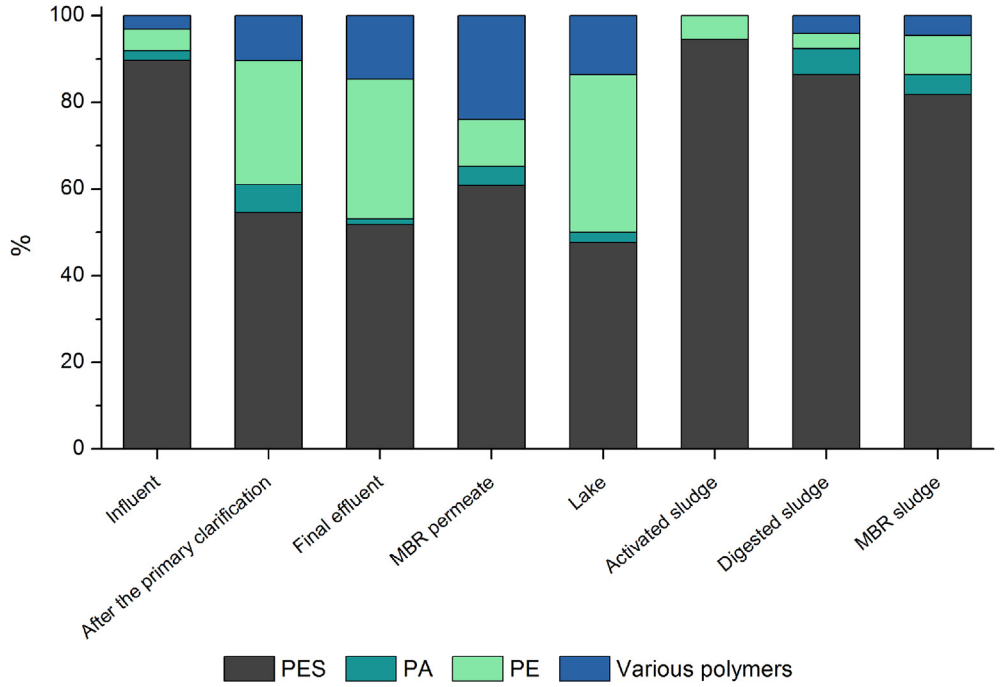

Fig. 1. The proportion of different polymers in different stages of the WWTP and a nearby lake. Various polymers include e.g. PE, PP and PES fragments with similar kind of appearance.

Table 1
Occurrence and description of polymers detected in different stages of the WWTP and recipient lake and identified by micro-FTIR and/or micro-Raman.

Polymer	Description	Percentage of the all microplastics collected
Polyester (PES), mostly polyethylene terephthalate (PET)	Fibers - Cross-section: Round, oval, flat - End: Cut, frayed, thickened - Appearance: Shiny or dull Particles - Shape: Flat, angular fragment - Hardness: Medium - Appearance: Shiny	79.1% Mostly fibers - Negligible proportion of MPPs
Polyethylene (PE)	Particles - Shape: Uneven flakes and fragments, spheres - Hardness: Medium to soft - Appearance: Dull or a bit shiny	11.4%
Polyamide, nylon (PA)	Fibers - Cross-section: Round, oval, flat - End: Cut - Appearance: Shiny	3.7%
Polypropylene (PP)	Particles - Shape: Uneven fragments - Hardness: Medium - Appearance: Dull	Negligible

cotton fibers with flat cross-section, and either twisted or straight appearance, were extracted from most of the samples. Also white or nearly transparent flat cellulose fibers, mainly originating from toilet papers, together with sand grains and glass fragments were also present in many of the samples. Altogether, from the initial 42 groups that were analyzed with either FTIR or Raman spectroscopy, 22 were excluded as they contained organic compounds (e.g. cotton, cellulose, chitin). For further studies, staining could be utilized in separation between plastics and other materials (Ziajahromi et al., 2017).

It is also worth noting that the spectra from FTIR and Raman analyses of most of the analyzed samples did not provide full match with the references, which could be caused by attached organic material, different additives used in plastics (Lenz et al., 2015) or the thermochemical treatments applied during the sample preparations. If some particle or fiber was not identifiable with available analysis, other representative from the same group was analyzed instead. Furthermore, the spectra libraries usually consist of spectra for pure substances, thus spectra obtained from environmental samples are expected to have low congruity compared to reference spectra (Murphy et al., 2016).

3.3. *Efficiency of WWTP to remove MPs*

In the current study, MPs concentrations varied between 0.1 and 124.7 MP/L in water samples and between 8.2 and 301.4 MPs/g dw in sludge samples. Highest average concentrations of microplastics were counted for influent samples and digested sludge; 57.6

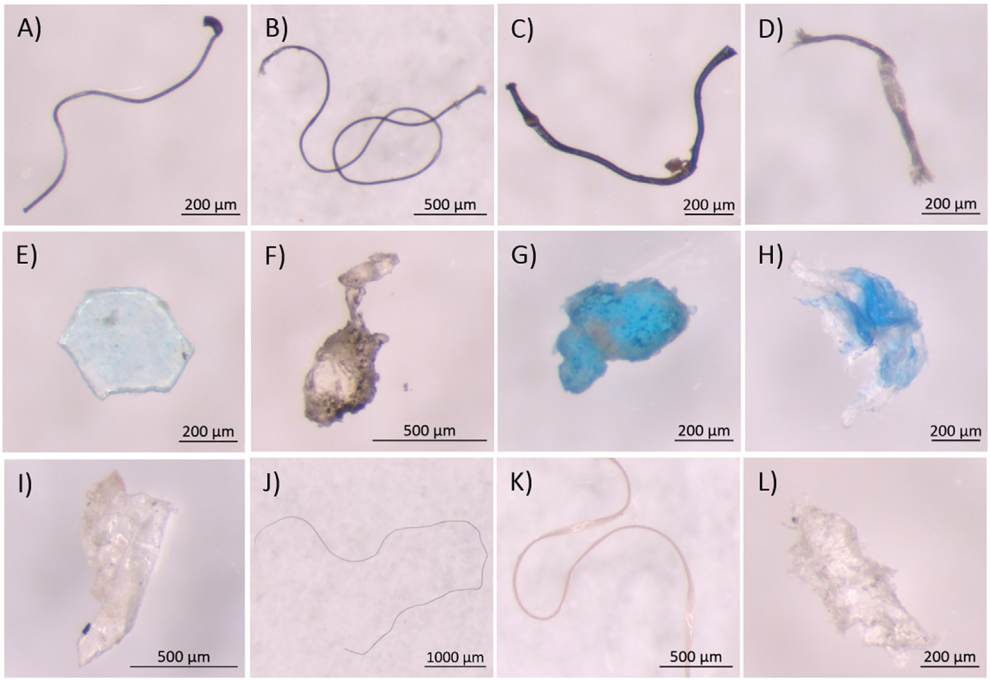

Fig. 2. Typical appearance of different polymers detected in different stages of the WWTP and recipient lake and identified by micro-FTIR and/or micro-Raman. (A–E) Polyester, (F–I) polyethylene, (J–K) polyamide and (L) polypropylene.

(±12.4) MP/L and 170.9 (±28.7) MPs/g dw, respectively (Table 2).

Reported MPs concentrations in WWTP effluents have varied between 0 and 91 MP/L (Table 3) (Browne et al., 2011; Carr et al., 2016; Dyachenko et al., 2017; Leslie et al., 2017; Magnusson and Norén, 2014; Mason et al., 2016; Michielssen et al., 2016; Mintenig et al., 2017; Murphy et al., 2016; Talvitie et al., 2015, 2017b; Ziajahromi et al., 2017). In this study final effluents had MPs concentration of 1.0 (±0.4) MP/L, on average, which is consistent with other previous reports (Browne et al., 2011; Ziajahromi et al., 2017). The large variation in previous results is mainly caused by different types of wastewaters and processes used in studied WWTPs (Mahon et al., 2017) together with different size limitations and sampling, preparation and identification methods (Underwood et al., 2017). Even though Ziajahromi et al. (2017) reported the decreasing concentrations of microplastics in WWTP effluents as the number of treatment steps is increased, most studies did not show similar trends. For example, one of the highest reported MPPs concentration, around 8.6 MPP/L, was reported by Talvitie et al. (2015) for effluent from the tertiary treatment.

Murphy et al. (2016) reported microplastic concentrations of approximately 2 MP in 2.5 g of centrifuged sludge. Talvitie et al. (2017a) reported 186.7 (±26.0) microlitter/g of dried sludge after centrifugation. They also found out that 20% of microlitter are returned to the wastewater flow via reject water. In the current study approximately 170.9 (±28.7) and 27.3 (±4.7) MP/g dw were counted for digested and MBR sludge, respectively. Noting, that our samples were collected before centrifugation and, in case of the MBR sludge, before any digestion.

According to the results of the present study, the Kenkäveronniemi WWTP had a MPs removal efficiency of 98.3% (Table 2). For microplastic particles and fibers the removal efficiencies were 89.8% and 99.1%, respectively. Removal efficiencies of total microplastics were in accordance with recent studies conducted in Lysekil, Sweden (99.9%) (Magnusson and Norén, 2014), the County of Los Angeles, USA (99.9%) (Carr et al., 2016) and Glasgow, UK (98.4%) (Murphy et al., 2016). Only one comparative research study of 7 WWTPs conducted in Netherlands reported removal efficiencies as low as 72% (Leslie et al., 2017).

In most related investigations, the majority of microplastics was

Table 2
Average concentrations (±SE) of microplastic particles and fibers in each stage of the wastewater treatment process in Kenkäveronniemi WWTP. The concentrations are presented in number per liter for water and per gram of dry weight for sludge.

Sampling point	Average concentration of microplastic particles (MPP/L or MPP/g dw) (±SE)	Average concentration of microplastic fibers (MPF/L or MPF/g dw) (±SE)	Average concentration of microplastics (MP/L or MP/g dw) (±SE)
Wastewater samples			
Influent	5.0 (±1.3)	52.6 (±11.3)	57.6 (±12.4)
Effluent from primary clarifier	0.2 (±0.1)	0.3 (±0.1)	0.6 (±0.2)
Final effluent	0.5 (±0.2)	0.5 (±0.3)	1.0 (±0.4)
MBR permeate	0.1 (±0.1)	0.2 (±0.1)	0.4 (±0.1)
Lake	0.2 (±0.0)	0.2 (±0.1)	0.3 (±0.1)
Sludge samples			
Activated sludge	1.3 (±1.3)	21.7 (±4.6)	23.0 (±4.2)
Digested sludge	9.8 (±5.7)	161.0 (±25.5)	170.9 (±28.7)
MBR sludge	3.3 (±2.4)	24.1 (±6.1)	27.3 (±4.7)

Table 3
Reported microplastics concentrations (MP/L) in final effluents of different WWTPs around the world.

Reference	MPs concentration (MP/L)	Lower size limit for fractionation (μm)	Type of the WWTP	Country
This study	1.05	250	Primary and secondary	Finland
Magnusson and Norén (2014)	0.00825	300	Mechanical, chemical and biological treatment	Sweden
Dyachenko et al. (2017)	0.02	125	Primary, secondary and tertiary	USA
Mason et al. (2016)	0.05	125	17 WWTPs	USA
Murphy et al. (2016)	0.25	65	Primary and secondary	UK
Carr et al. (2016)	0	45	Primary, secondary and tertiary	USA
Ziajahromi et al. (2017)	0.28	25	Primary, secondary and tertiary	Australia
Ziajahromi et al. (2017)	0.48	25	Primary and secondary	Australia
Ziajahromi et al. (2017)	1.54	25	Primary	Australia
Michielssen et al. (2016)	0.5–5.9 (incl. all textile fibers)	20	2 WWTPs: primary, secondary and tertiary	USA
Mintenig et al. (2017)	0.1–10.05	20	12 WWTPs: mostly secondary and tertiary	Germany
Talvitie et al. (2015)	13.5 (incl. all textile fibers)	20	Primary, secondary and tertiary	Finland
Talvitie et al. (2017b)	0.005–0.3	20	4 tertiary WWTPs	Finland
Leslie et al. (2017)	9–91	0.7	7 WWTPs	Netherlands
Browne et al. (2011)	1	- (filtered)	Primary, secondary and tertiary	Australia

removed in the beginning of the wastewater treatment process during mechanical and chemical pretreatments, solids skimming and sludge settling processes (Carr et al., 2016; Talvitie et al., 2017a). Therefore, effluent filters were found to have only minimal effect on the removal of MPs from wastewaters (Carr et al., 2016). As far as the present study is concerned, 99.0% of MPs were removed already before the aeration process, which is consistent with other studies involving CAS process. In addition, it was found that MBR technology was more efficient (99.4%) in removing MPs from wastewaters compared to the CAS process. On average, MBR permeate contained lower concentrations of microplastics (0.4 ± 0.1 MP/L) than the final effluent from the main CAS process (1.0 ± 0.4 MP/L).

The average inflow of wastewater during sampling days was 10 766 (±501) m^3 and 10 540 (±425) m^3 of final effluent and 87.3 (±5.5) m^3 of digested sludge were released. Consequently, in the light of MPs concentrations, approximately 1.0×10^7 and 4.6×10^8 microplastics were discharged daily with the final effluent and digested sludge from the Kenkäveronniemi WWTP, respectively. When comparing the amount of discharged MPs to the amount of MPs channeled to the plant with the influent flow (6.2×10^8 MPs/d), a fraction of the microplastic flux (1.5×10^8 MPs/d) seems to be trapped within the WWTP. This could be related to their possible entrapment with solid materials from grit separation and grease from primary clarification (Murphy et al., 2016).

Although the wastewater treatment process is basically a linear concept, nonetheless some recycled flows do exist within WWTPs. These flows include reject water from the sludge dewatering, water from rinsing of removed solids and grease, floating materials from the primary sedimentation and waste activated sludge (WAS). All of these flows are concentrated in the beginning of the process and, thus, collected influent samples consist of both incoming screened wastewater and “returning” flows. The main remark in this regard is that the results of this study do not represent the differences between raw influent and treated effluent, but differences of the MPs concentrations between different stages of the WWTP process and ultimately the MPs concentrations discharged with the final effluent.

As for the lake water samples, collected from nearby the effluent outlet, the analysis revealed microplastics concentrations of 0.3 (±0.1) MP/L. Previously, only 0.0008–0.003 MP/L were counted from the surface waters of two Italian lakes, when using mesh size of 300 μm (Fischer et al., 2016). In comparison with the MPs concentrations reported for other water bodies nearby the discharge of WWTP effluents, the concentrations of the present study were considerably higher. For example, MPs concentrations of 0.001–0.002 MP/L were reported for seawater collected 20–200 meters from the WWTP effluent outlet in Sweden, using mesh size of 300 μm (Magnusson and Norén, 2014). In current study, the lake water was sampled only to estimate the amount of MPs in close proximity to the discharge location of treated wastewater. Hence, the reported concentrations are neither representing the average MPs concentration in the recipient lake, nor being correlated with the one discharged from the WWTP. Other possible sources of MPs in lake water include e.g. stormwater, littering and atmospheric fallout (Dris et al., 2015), which are not further discussed here.

Overall, microplastics concentrations were slightly higher in final effluent than in the effluent from primary clarifier, especially for microplastic particles. This might be caused by the more turbulent flow of the effluent from primary clarifier compared to the more stable flow at the sampling point of the final effluent. Besides, since most of the particles were polyethylene and they are normally floating in water (density 0.92–0.97 g/cm^3) (Hidalgo-Ruz et al., 2012), particles might have concentrated to the upper layer of the final effluent flow, where samples were collected.

3.4. Occurrence and distribution of MPs

3.4.1. Distribution by type

Fig. 3 clearly shows that MPFs were most abundant types of microplastics in the wastewater and sludge of the studied WWTP. For instance, the highest occurrence of MPFs in relation to MPPs was in the sludge fractions with highest occurrence of 94% in the activated sludge and then in the initial effluent (92%). Even at the lowest, the fibers were accounted for 53% and 50% of all counted MPs in final effluent and lake water, respectively. The same tendency, i.e. predominance of MPFs over MPPs, was also reported in the literature for both wastewater effluents (Leslie et al., 2017; Mason et al., 2016; Michielssen et al., 2016; Talvitie et al., 2015; Ziajahromi et al., 2017) and the environment (Browne et al., 2011).

Since there are hardly any plastic industry in the studied area and no stormwater runoffs are conducted to the WWTP, most of the microplastics are originated from households and different municipal services. In this context, the high amounts of plastic fibers indicates the high impact of laundry and textile handling activities on the microplastic emissions. Another, less studied route of MPs to the WWTP could be infiltration of sewer system.

3.4.2. Size distribution

As shown in Fig. 4, 64% of all recovered microplastics from the samples were smaller than 1 mm, half of which were also smaller than 0.5 mm. Talvitie et al. (2017a) reported that depending on the studied stage of the WWTP process, only 20% of the collected microlitter were larger than 300 μm. The difference is due to the

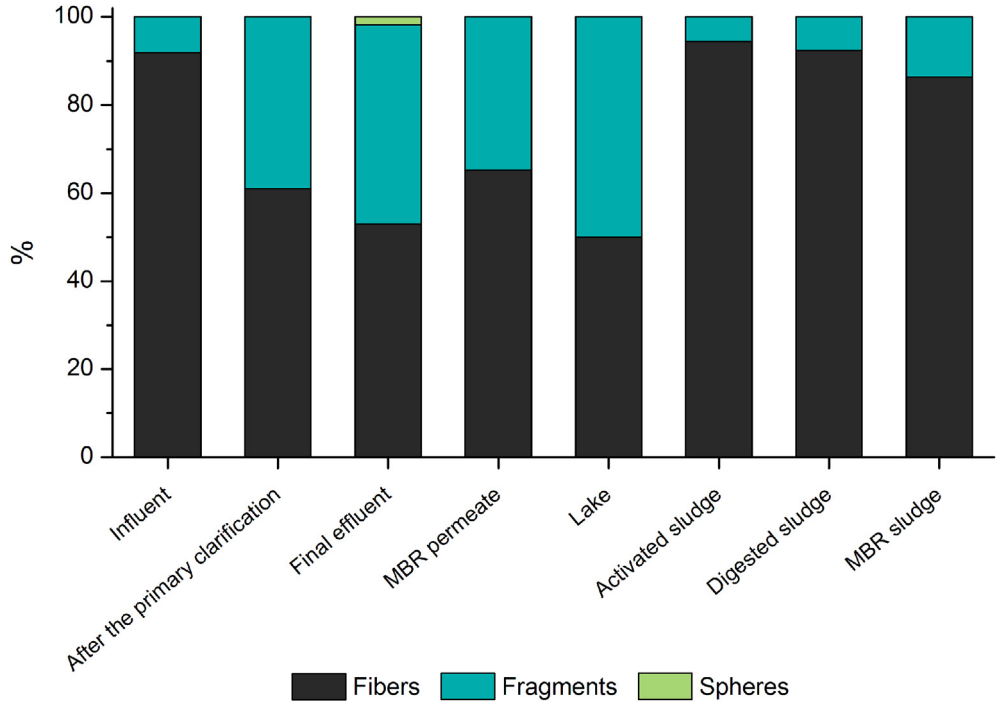

Fig. 3. The proportion of different types (fibers, fragments, spheres) of microplastics in different stages of the WWTP and a nearby lake.

different sampling methods, since they collected MPs with smaller mesh size (20 μm) than was used in this study (250 μm). Consequently, if smaller sized MPs would have been caught with finer mesh sizes similarly in the current study, the amount of recovered MPs would have been higher.

In present study, the main size of MPs in final effluent is between 0.5 and 1 mm. Although MBR process is more efficient than CAS to remove microplastics, the proportion of the smallest-sized MPs (<0.25 mm) is at its highest value in MBR permeate (Fig. 4). Equally, about one fourth of the MPs collected from lake samples

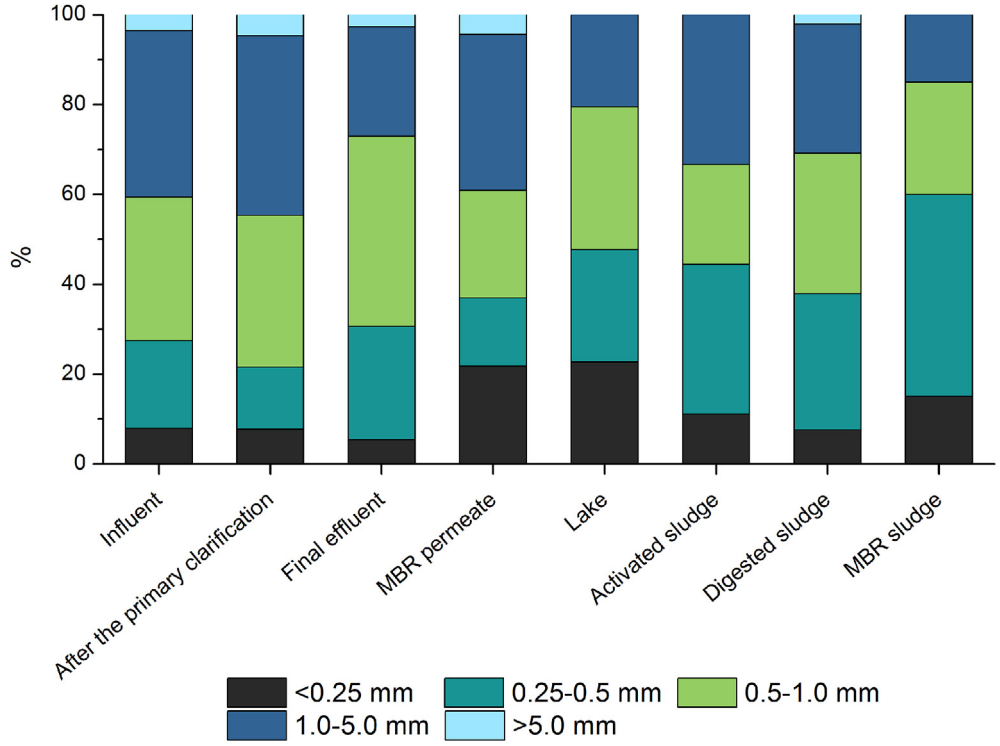

Fig. 4. The proportion of different sizes of microplastics in different stages of the WWTP and a nearby lake. Size was measured as the fiber length and the largest dimension of the particle.

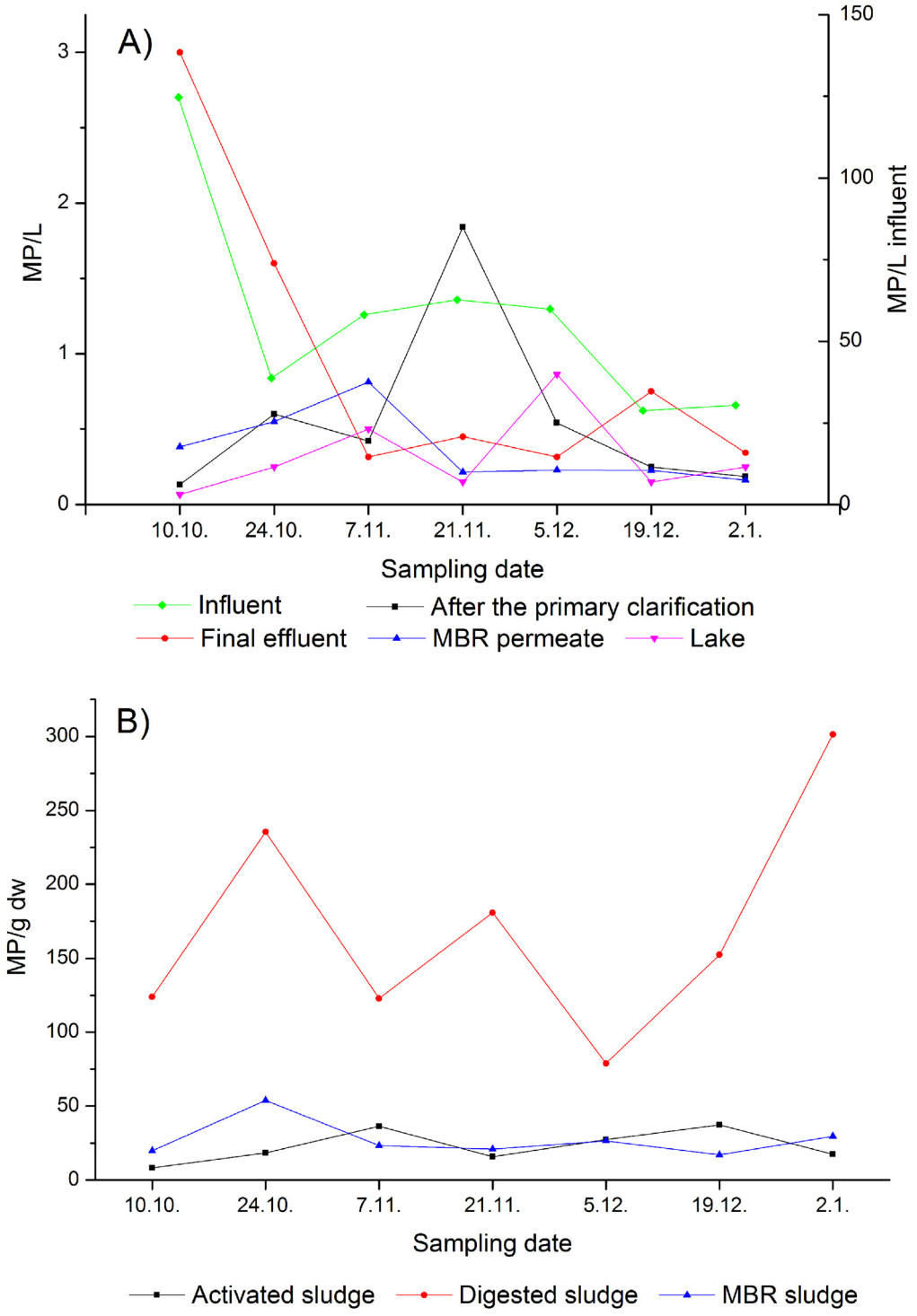

Fig. 5. Microplastic concentrations in (A) water (MP/L) and (B) sludge (MP/g dw) in different stages of the WWTP process at different sampling dates. The results for the influent samples in (A) have their own scale in right Y-axis.

were smaller than 0.25 mm, which highlights the importance of including smaller sized MP fractions to future studies. Large MPs in MBR permeate might have caused by contamination in the open tank, from where permeate was collected.

Because the dimensions and shapes of fibers and some particles are not symmetrical, a fraction of MPs might pass through the mesh of the sieves even though one of their dimensions would be longer than the mesh size. As a result the number of MPs (mostly fibers) counted from the samples reflects only the retained fraction of the microplastics (Magnusson, 2014). On the other hand, samples might also have fibers and particles with smaller dimensions than the mesh size, if they are entangled together or attached to larger particles while sieving (Castro et al., 2016). Overall, the risk of losing small-sized MPs is higher if 250-μm mesh size is used, and the proportion of small microplastics would be much larger if smaller mesh size would have been used (Klein et al., 2015; Ziajahromi et al., 2017).

3.4.3. Spatiotemporal variations of MPs concentrations

Microplastics concentrations varied substantially during the sampling campaign in both water and sludge samples (Fig. 5), with similar patterns when concentrations of MPFs and MPPs were observed separately. Therefore, the MPs concentrations in WWTPs reported for single sampling events do not give reliable set of data, based on which the microplastics pollution issue could be properly assessed and dealt with. In order to collect more representative samples and including diurnal variation in the approximation of microplastic concentrations in WWTP, automatic composite sampling could be used (Talvitie et al., 2017a).

Up until now, most research studies were conducted within a timeframe of days. Nonetheless, few other studies pointed out this high variation of MPs concentrations in wastewaters within weeks and seasons (Mintenig et al., 2017; Talvitie et al., 2017a). The sampling campaign of this investigations was conducted between the second week of October and the first week of January in order to assess the occurrence of MPs in wastewaters during the autumn and winter seasons in Nordic environment. Based on the presented results of this study, the authors emphasize the need to extend the monitoring campaign to cover the spring and summer seasons in order to have an estimate of the annual variation of MPs in wastewater and the respective ability of WWTPs to effectively deal with such seasonal variation.

In addition to the seasonal variation, there might be diurnal variation in the MP concentrations in the influent. This was not taken into account in the sampling and calculations of the removal efficiency of MPs in the studied WWTP, which might have added some uncertainty in the reported results. Thus for long-term sampling campaigns, considering the hydraulic retention time in different process parts should be taken into account for more reliable assessments.

4. Conclusions

The results of the 3-month sampling campaign show that 98.3% of incoming microplastics were removed during the treatment process of the studied WWTP. According to the MPs occurrence data, microplastic fibers (82%) are posing a more severe problem than microplastic particles (18%), hence the need to put more focus on the occurrence and fate of MPFs in wastewaters and receiving water bodies.

Furthermore, this study contributed to the research effort assessing the occurrence and temporal variations of MPs within WWTPs and the efficiency of conventional activated sludge process and advanced MBR technology to remove microplastics. In this regard, it was found that MBR process had a slightly better removal efficiency of microplastics (99.4%) compared to the overall CAS-based process (98.3%). Of all analyzed polymers, polyester (mostly PET) was revealed to be most abundant in collected samples by constituting 79% of the entire MP load and being present in all studied stages of the WWTP.

The high variation in the determined MPs concentrations highlights the importance of conducting several sampling events, when studying microplastics in media of variable contents.

In the current study, grab sampling covering separated time frames, sampling locations and contamination might have caused uncertainties to the reported results. Overall, in this field of microplastic research, standardized protocols have to be developed, because of the high amount of uncertainties during sampling, sample treatment and characterization, and in order to provide reliable methods and generate comparable set of data between researchers.

Another important issue is the urgent need for more reliable ways of identifying plastic particles and fibers from non-plastics, especially with increasing quantities of identified particles and fibers. The applicability of staining and imaging database of confirmed microplastics have already been studied, but more research should be conducted to optimize them and find out the most suitable and effective method for microplastics identification.

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Conflicts of interest

None.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.watres.2018.01.049.

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