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Treatment characteristics of microplastics at biological sewage treatment facilities in Korea

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ABSTRACT

Microplastics that are contained in household dust, personal care products, and other factors, are discharged into sewage treatment facilities (STF). While these microplastics are treated at the STF with a high treatment efficiency through settling, precipitation, filtering, and other treatments, considering the large amount of effluent, large quantities of microplastics are still discharged into marine environments. In this study, biological STF using the anaerobic-anoxic-aerobic (A2O), sequence batch reactor (SBR), and the Media processes were investigated to confirm the efficiency of these treatments and the associated amounts of microplastics released for each process. The three investigated processes were found to have treatment efficiencies of about 98% or more. However, due to the large amount of effluent, more than four billion pieces of microplastic were released each year in each facility. Thus, even though biological STF show high treatment efficiencies, substantially large amounts of microplastics are still released into the marine environment.

1. Introduction

Plastics Europe reported that the worldwide production of plastics in 2005 was about 230 million tons, and production has since increased steadily, with about 320 million tons of plastic being produced in 2015 (Plastics Europe, 2016). It is assumed that the production of microplastics also increased accordingly. Plastics smaller than 5 mm are defined as microplastics, and are classified into primary and secondary microplastics, depending on how they were generated (Masura et al., 2015). Primary microplastics contained in cosmetics, blasting media, cleaning products, personal care products, and so on, are artificially manufactured from raw plastic material (Magnusson et al., 2016; Masura et al., 2015). Secondary microplastics are made by the mechanical degradation, photo degradation, bio degradation, thermo-oxidative degradation, hydrolysis, and other mechanisms, of larger plastics in the environment (Andrady, 2011; Magnusson et al., 2016; Masura et al., 2015). Van Sebille et al. estimated an amount of from 15 to 51 trillion and from 93 to 236 million tons of microplastics in the world's oceans (van Sebille et al., 2015). However, if considering the amounts of microplastics that are ingested by marine organisms, that settle on the seabed, and that attach to the sand on beaches, the actual amount will be larger than van Sebille's estimate.

Due to high specific surface area and hydrophilicity compared to meso and macro plastics, microplastics adsorb and desorb in their surface persistent organic pollutants (POPs), such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and metals (Ashton et al., 2010; Bakir et al., 2014; Cole et al., 2011; Frias et al., 2010). Plastics also contain additives such as polybrominated diphenyl ethers (PBDEs), phthalates, bisphenol A, and so on (Cole et al., 2011). Diphenyl ether, phthalate, and bisphenol A are known as endocrine disruptors that can mimic, compete with, and disrupt the synthesis of endogenous hormones (Cole et al., 2011). These microplastics can exhibit a random distribution within a water depth in marine environments by changing the density of the plastics by adjusting the biofilm coating (Carr et al., 2016).

Microplastics discharged into the environment include microbeads used in personal care products, fibers produced from indoor dust and washing, synthetic rubber from tire fragments, road and building paint from weathering, blasting media, and so on (Lee and Kim, 2017b; Sundt et al., 2014; Magnusson et al., 2016). Among these microplastics, microbeads used in personal care products and fibers generated by laundering eventually flow into sewage treatment facilities (STF). Furthermore, in STF equipped with combined sewerage, microplastics from tire fragments on the road are washed away by rain, and are eventually deposited into the sea. In July 2017 in Korea, microplastics used as raw materials in cosmetics such as bathing and cleaning products were forbidden according to the 'Regulations on Safety Standards for Cosmetics' (National Law Information Center, 2017c). However, such

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cosmetic products manufactured prior to this regulation continue to be sold today (Ministry of Food and Drug Safety, 2016). Therefore, despite the prohibition, microplastics continue to discharge into STF from personal care products. In 2015, the total sewerage installation extension in Korea was approximately 140,000 km, of which 44,000 km (32%) was combined sewerage (Ministry of Environment, 2016). While this combined sewerage length has decreased by about 5000 km since 2006 (Ministry of Environment, 2016), a considerable amount of combined sewerage installed. In Korea, biological sewage treatment methods are mostly adopted. The types of public STF with a capacity of 500 m^3/day or more, as of 2013, are categorized as follows: 23.7% use the anaerobic-anoxic-aerobic (A2O) process, 34.8% use the sequence batch reactor (SBR) process, and 22.8% use the Media process (Korea Environment Corporation, 2015). The above three processes account for > 80% of the public STF in Korea, while membrane bioreactor (MBR), long term aeration, and special microbial processes account for the remaining percentage (Korea Environment Corporation, 2015). The A2O process consists of an anaerobic, anoxic, and aerobic basin. The anaerobic, and anoxic basin serves as the release of phosphorus, and as the denitrification, respectively (Kang et al., 2008). The aerobic basin serves as the nitrification, luxury uptake of phosphorus, and decomposition of organic matter (Kang et al., 2008). The SBR process has four phases that are a fill phase, react phase, settle phase, and decant phase in one reactor (Kang et al., 2008). The react phase dedicate the alternating aerobic and anoxic cycles (Kang et al., 2008). The Media process forms an anaerobic, anoxic, and aerobic basin and uses filled carrier (Korea Environment Corporation, 2015).

The purpose of this study is to compare the microplastics treatment efficiencies of biological STF (A2O, SBR, and Media process) which are widely used in Korea. Also, the types and sizes of the microplastics discharging into STF equipped with combined sewerage are analyzed and these are provided as basic data for determining ways to reduce the amount of microplastics discharged into marine environment. Meanwhile, Magnusson and Noren (2014) reported that the concentration of microplastics larger than 300 μm in effluent at a sewage treatment facility was 0.008 particles/L, showing 99.9% of removal efficiency, Murphy et al. (2016) reported that the concentration of microplastics of larger than 65 μm in effluent at another sewage treatment facility was 0.25 particles/L, showing 98.4% of removal efficiency.

2. Materials and methods

Table 1 shows the location, facility capacity, treated area, sampling date, and total precipitation from July to September of the investigated STF. The A2O, SBR, and Media processes were sampled at the STF of M-city, Y-city, and S-city in Korea, respectively. The facility capacity, size, and population of the treated area ranged in the descending order of Media, SBR, and A2O types of processes (National Sewerage Information System, 2018; KONETIC, 2015a,b,c). Precipitation in Korea is concentrated from June to September (Kang, 2000; Korea Meteorological Administration, 2018), and sampling was conducted twice from July to September. Figs. 1–3 show the sampling points of the STF being investigated process. A schematic diagram of the A2O process was obtained from the KONETIC website (KONETIC, 2008). A schematic diagram of the SBR process was confirmed by the Information Open Portal (2017). A schematic diagram of the Media process was obtained from person in charge. Influent and effluent were sampled at 10 L before the coarse screen and 100 L in the UV sterilization tank, respectively. They were then sieved in the field with a 106 μm mesh sieve, then stored in a zipper bag and transported to the laboratory. The influent and effluent were sampled separately to check the suspended solids (SS). The sludge cake at the dehydrator was placed into a zipper bag using a shovel and then transported to the laboratory. The sampling points including the sludge cake were selected to determine the mass balance of microplastics.

The moisture, combustibles, and ash of the sludge cakes transported to the laboratory were analyzed based on the 'Official test standard of waste' in order to identify combustibles component and microplastics (National Law Information Center, 2017a). To compare the treatment efficiencies of STF for microplastics and SS, influent and effluent were analyzed for SS based on the 'Official test standard of water pollution' (National Law Information Center, 2017b).

Analysis of microplastics for the samples was carried out based on the National Oceanic and Atmospheric Administration (NOAA) (Masura et al., 2015). Lee and Kim (2017a) reported that sieve sorting would be appropriate following initial density separation to reduce the solids content of the sludge cake. However, due to the characteristics of the agglomerated sludge cake, this study decomposed the sludge cake through organic decomposition. Samples of influent, effluent, and sludge cake transferred to the laboratory were placed in a beaker for the decomposition of organic matter. 20 mL of 0.05 M Fe (II) solution and 20 mL of 30% hydrogen peroxide ($H_2O_{2)}$ solution were then added and allowed to stand for 5 min. After standing, stirring (80 rpm), and heating (75 °C), the samples were removed from the beaker and placed on a hot plate for 30 min. The conditions such as temperature and time are determined by basis on NOAA (Masura et al., 2015). Since this temperature can damage microplastics, it is necessary to pay attention to temperature adjustment (Munno et al., 2018). When bubbles are generated, the heating must be stopped immediately, and a small amount of distilled water is added to avoid boiling of the decomposition liquid. Samples were classified by size using 106 μm and 300 μm mesh sieves. The size of 106 μm was selected considering the average value of 20–200 μm, which is the size range of microzooplankton (Yang and Choi, 2003), and the size of 300 μm was selected considering the general study of plankton and floating waste (Arthur et al., 2009).

Yu et al. (2016) reported that when three repeated density separations were applied using a saturated sodium chloride (NaCl) solution with a density of about 1.27 g/mL, microplastics were recovered at a rate of > 90% in the sand matrix. However, in previous research in this field, the solid matter was stored in a petri dish using a small amount of distilled water without density separation because the efficiency of density separation between influent and effluent was insignificant. The sludge cake was transferred to a separatory funnel using a small amount of distilled water. Density separation was then carried out using a zinc chloride (ZnCl2) solution having a density of about 1.6 g/mL. The samples were sieved and then stored in a petri dish with a small amount of distilled water.

The samples stored in the petri dish were visually classified and their sizes, colors, and shapes were confirmed using a HT003 USB digital microscope (HiMax Tech, Korea). For microplastics with sizes of 106–300 μm and > 300 μm, monitor magnifications of about 320 and 110 magnification were used, respectively. After observation, black pieces suspected of being tire fragments were confirmed through the

Table 1
Facility capacity, sampling dates, and precipitation for each site of each process.

Process	Site	Facility capacity	Treatment area	Population in treatment area	1st sampling date	2nd sampling date	7–9 months precipitation
A2O	M-city	35,000 m^3/day	346 ha	67,700	2017. 07. 05.	2017. 08. 08.	449.4 mm
SBR	Y-city	110,000 m^3/day	1770 ha	235,711	2017. 07. 10.	2017. 09. 12.	589.0 mm
Media	S-city	130,000 m^3/day	5044 ha	245,200	2017. 07. 14.	2017. 09. 27.	475.3 mm

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