



Wastewater treatment plant effluents as source of cosmetic polyethylene microbeads to freshwater



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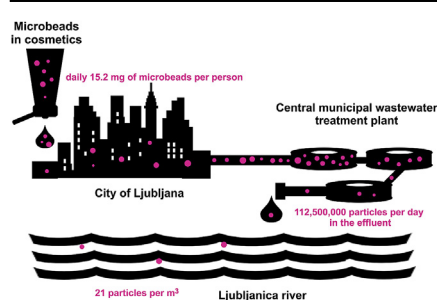
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HIGHLIGHTS

- Polyethylene microbeads are present in cosmetic products.
- 15.2 mg per person per day of microbeads are released into sewage system.
- Smaller particles (up to 70 μm) are captured within activated sludge during biological treatment.
- Microbeads concentrations in river can reach 21 particles/m³.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastics in the environment are either a product of the fractionation of larger plastic items or a consequence of the release of microbeads, which are ingredients of cosmetics, through wastewater treatment plant (WWTP) effluents. The aim of this study was to estimate the amount of microbeads that may be released by the latter pathways to surface waters using Ljubljana, Slovenia as a case study. For this purpose, microbeads contained in cosmetics were in a first step characterized for their physical properties and particle size distribution. Subsequently, daily emission of microbeads from consumers to the sewerage system, their fate in biological WWTPs and finally their release into surface waters were estimated for Ljubljana. Most of the particles found in cosmetic products were <100 μm. After application, microbeads are released into sewerage system at an average rate of 15.2 mg per person per day. Experiments using a lab-scale sequencing batch biological WWTP confirmed that on average 52% of microbeads are captured in activated sludge. Particle size analyses of the influent and effluent confirmed that smaller particles (up to 60–70 μm) are captured within activated sludge while bigger particles were detected in the effluent. Applying these data to the situation in Ljubljana indicates that about 112,500,000 particles may daily be released into the receiving river, resulting in a microbeads concentration of 21 particles/m³. Since polyethylene particles cannot be degraded and thus likely accumulate, the data raise concerns about potential effects in aquatic ecosystems in future.

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1. Introduction

Since the 1970, pollution of marine ecosystems by small pieces of plastic materials has been recognized as a serious environmental problem (Carpenter and Smith, 1972; Lusher et al., 2015). These

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plastic fragments, commonly referred to as microplastics, are characterized by a size below 5 mm (Andrady, 2011). In recent years microplastics were put in a more systematic context, where it was suggested to separate industrially manufactured microparticles (e.g. abrasives in cosmetic products) – considered as primary microplastics – from particles produced through fragmentation of larger plastic items (Fischer et al., 2015). Microplastic pollution is of truly global concern as it has been reported for coastal areas (Ng and Obbard, 2006) and open ocean (Law et al., 2010) from the poles to the equators including remote habitats (Derraik, 2002; Ivar do Sul and Costa, 2014). Moreover and driven by their high diversity in terms of physicochemical properties, microplastics occur in divers compartments within one ecosystems; they accumulate in sediments (Dekiff et al., 2014), disperse in the water body or float on the water-air interface (Andrady, 2011).

In contrast to marine ecosystems, information on microplastic pollution in freshwaters is limited. Some recent studies demonstrated that freshwater ecosystems are carrying similar loads as marine systems. Zhang et al. (2015), for instance, reported concentrations of floating microplastics in China of up to 13,617,500 particles/km² (above 0.112 mm). Eriksen et al. (2013) found lower levels (0.355–0.999 mm, 736,749 particles/km²) in the Laurentian Great Lakes, which was linked to wastewater treatment plant (WWTP) effluents. A number of further studies investigated to which extend WWTP effluents may indeed be considered as major pathway for microplastics to enter the freshwater environment. Results are, however, often inconsistent, which may be explained by the differences in sampling, identification and enumeration techniques as well as expression of results (Eerkes-Medrano et al., 2015). Van Cauwenberghe et al. (2015), for example, reported an overall removal efficiency of a mechanical biological WWTP in Belgium of 64% and 18% for microplastic particles and fibres, respectively. Murphy et al. (2016), on the other hand, documented a removal efficiency of 98% in a mechanical biological WWTP in Glasgow, whereas Carr et al. (2016) suggest a nearly complete removal after tertiary treatment. Although these investigations give a rough idea about the removal efficiency for all types of plastic particles found in wastewaters (microbeads, textile fibres), polyethylene microbeads as used in cosmetic products have not specifically been assessed.

The present study, hence, aims at estimating the quantity of microbeads that can be released from cosmetic products daily, their fate in WWTP and subsequently their emission *via* WWTPs to surface waters. In a first step, however, the present study characterized physical properties and the particle size distribution of microbeads as present in commercial cosmetics.

2. Material and methods

2.1. Analysis of microbeads from cosmetic products

Five microbeads-containing products (three body and two facial scrubs) that are used at least on a weekly basis have been purchased from two international drugstores and one direct-selling company. All products are manufactured by leading cosmetic companies and can be purchased worldwide at a relatively low price (<6 €). The products A and C were facial scrubs, while B, D and E were body scrubs. Product E was obtained from the direct-selling company. All products have been purchased during the years 2015 and 2016.

To extract microbeads, about 20 mL of each cosmetic product were slowly added to warm deionized water (~50 °C) and stirred in 1000 mL beaker on a magnetic stirrer set at 400 rpm until the product was completely dissolved. The solution was subsequently filtered over Whatman™ filter paper (pore size 4–12 μm). Retained microbeads were several times washed with deionized water to

remove potentially remaining ingredients of cosmetic products. Microbeads were subsequently carefully washed from the filter into glass beakers and dried overnight at 60 °C.

To determine the particle material, IR spectra were recorded on a Spectrum 1000 FT-IR spectrophotometer (Perkin Elmer) incorporation the samples in KBr discs (2 mg of sample was pulverized with 98 mg of KBr pressed into a KBr disc).

Particle size was analyzed by laser diffraction analyzer S3500 (Microtrac). Therefore, 0.5 g of microbeads was dispersed in demineralized water containing 0.1% sodium dodecyl sulfate to reduce microbead aggregation. The dispersion was slowly transferred to the sampling cell. The particle size distribution of each product was determined in triplicate. The number of particles per g of sample was calculated using the numerical size distribution obtained by the Microtrac particle sizer. The numerical size distributions represent the percentage of particles in each size class based on particle numbers – the summed percentages of all classes is thus equal to 100 for what is assumed to represent 100 particles. First, the mass of particles in each size class was calculated assuming spherical polyethylene particles with constant density. The final result was obtained as the quotient of the number of particles and the total weight of 100 particles (Table 1).

Single point BET (Brunauer-Emmett-Teller) specific surface area ($S_{\text{single point}}$) of samples were determined using an ASAP 2020 instrument (Micromeritics) (ISO, 2010b). BET method determines the amount of adsorptive gas required to cover the accessible surface of microbeads at a constant temperature and serves as a proxy of the surface area (Turner and Holmes, 2011; Hu et al., 2017).

Morphology of microbeads was observed by a field emission scanning electron microscope (SEM) Supra 35VP (Carl Zeiss). All observed samples were mounted on aluminium stub and fixed with carbon tape. Particles were coated with thin layer of silver and palladium alloy. Samples were imaged at a 1-kV accelerating voltage.

All results are expressed as mean values (MN) together with standard deviation (\pm SD). Also, the number of replicates (n) is detailed.

2.2. Lab-scale wastewater treatment plant

The operational parameters of the lab-scale WWTP were set up according to technological data of Central Municipal Wastewater Treatment Plant (CMWTP) in Ljubljana, Slovenia (Vodovod-Kanalizacija, 2017). CMWTP is a single-stage mechanical and biological treatment plant intended for the removal of undissolved substances, carbon and nitrogen compounds. The primary treatment is composed of coarse and fine screens with consequent basins for removal of grease and sand. In the lab-scale WWTP the mechanical treatment was not included and microbeads removal efficiency of this stage was estimated based on literature data: Van Cauwenberghe et al. (2015) observed a roughly 20% removal of a diverse set of microplastics by screens. Similarly, Talvitie et al. (2015) reported microplastics removal efficiency of just above 30% following screens, grit removal, pre-aeration and primary sedimentation. Those results are in accordance with Magnusson et al. (2016) who observe negligible retention of microplastics in a WWTP equipped with a primary sedimentation tank and screens only. These insights suggest that a removal efficiency during primary treatment of approximately 25% as reasonable. The microbeads removal efficiency of the biological treatment was simulated by a lab-scale sequencing batch reactor (SBR). During the SBR process similar condition as in the CMWTP proceeded – a first part of the biological treatment at the CMWTP aimed for reaction between activated sludge and wastewater. The subsequent settling tank separates treated wastewater and activated sludge. Both

Table 1

Concentration, specific surface area, particle size distribution, 10th and 90th percentile of particle size distribution and mean number of particles per mg of microbeads from two facial scrubs (A, C) and three body scrubs (B, D, E).

Parameter	Product				
	A	B	C	D	E
	Facial scrub	Body scrub	Facia scrub	Body scrub	Body scrub
Concentration of microbeads g per 100 mL of the product	0.42	2.47	1.06	0.87	11.12
Specific surface area $S_{single\ point}$ (cm ² /g)	<1	10	33	295	189
Mean number particle size distribution (μm) (mean ± SD, n = 3)	37.66 ± 16.79	71.30 ± 34.29	55.76 ± 28.88	95.95 ± 68.99	74.95 ± 36.25
10th percentile of particle size distribution (μm)	16.88	31.04	24.38	37.76	32.86
90th percentile of particle size distribution (μm)	68.99	128.40	104.40	202.30	132.70
Mean number of particles per mg of microbeads ^a	3108	853	2185	625	1186

^a Calculated values.

processes are also covered during the SBR process. According to Abdelkader (2009) the SBR process is strongly comparable to conventional biological treatment plants when based on the reduction of the biochemical oxygen demand (BOD) and total suspended solids (TSS). SBR is considered more efficient for the removal of ammonium nitrogen (~10% more efficient). Furthermore concentration of activated sludge and the hydraulic retention time (HRT) were set according to technological data of the CWWTP ensuring comparable contact time between microbeads and activated sludge flocks in the aeration basin of the biological treatment stage.

For the laboratory experiments, three glass cylindrical reactors with a total volume 5 L were used. One of them was fed with synthetic sewage only representing an uncontaminated control (without microbeads). Furthermore, two independently working reactors labelled MP1 and MP2 were used to evaluate microbeads removal efficiency. They were also fed by synthetic sewage, but 0.5 g/L of microbeads were introduced into each reactor at each cycle. Only microbeads from product B were used, because product B was indicated by consumers as the most often used body scrub (see the information about consumer survey below). Standard synthetic sewage (ISO, 2004) was used instead of real sewage to limit variations. It was prepared from inorganic salts and 160 mg/L of peptone that gives mean DOC concentration in the influent of about 60 mg/L (ISO, 2004). Activated sludge also originated from the CWWTP, it was collected from the aeration basin, thickened by gravity settling and immediately transported to the laboratory. In the laboratory, activated sludge was several times washed with tap water to remove coarse particles and remaining wastewater. Following a final settling step, its concentration was determined. It was consequently diluted in glass reactors into final concentration 3.1 ± 0.1 g/L, reflecting the situation at CWWTP.

The SBR operation parameters were: 12 h of aeration (on magnetic stirrer (200 rpm) with aeration from the bottom via a silicon rubber tube) and 8 h of settling (HRT = 20 h). An additional 2 h were used for the loading and withdrawal, respectively. Thus, each cycle was completed after 24 h and proceeded 6 times to assess for variation in the microbeads removal efficiency. Microbeads in the effluent were washed several times with diluted sulphuric acid (~5% v/v) to remove activated sludge flocks. Subsequently, particle size distribution of microbeads in the effluent was measured (description is given in chapter 2.1). To evaluate an efficiency of the SBR system in comparison to CWWTP in terms of heterotrophic and nitrification activity the dissolve organic carbon (DOC) (ISO, 2010a) and ammonium nitrogen (N-NH₄⁺) (ISO, 1984) concentration prior and after treatment were quantified in all reactors.

2.3. Estimation of daily emission of microbeads to the sewerage system

To extrapolate from the SBR study to the potential input of

microbeads into the receiving stream, the city of Ljubljana, Slovenia with 300,000 inhabitants was selected. The CWWTP (360,000 population equivalent) treats about 80,000 m³ of wastewater daily. The treated effluent is released into Ljubljanica River, where it is diluted by a factor of approximately 70.

For this exercise, the amount and frequency of application of body and facial scrubs by the local population was estimated involving a consumer survey. The survey was performed in the vicinity of Ljubljana, Slovenia by direct interviews of consumers on streets, in faculties, and on work places. Additionally, the present study took advantage of e-mails and social networks asking for completion of an online survey (Google Forms). The survey was anonymous and the only data collected related to costumers were age, sex and confirmation that they are residents of Ljubljana or live in its vicinity (30 km). Respondents were asked to check their personal care products and provide type of exfoliator in their facial and body scrubs. They were also asked to indicate the brand of the cosmetic product allowing to back check if consumers listed the type of exfoliator correctly. To estimate the daily emission of polyethylene microbeads from body and facial scrubs, respondents provided information about frequency and quantity of the product usage (evaluated by the frequency they press the package). Finally, it was prompted whether the respondents actively chose for a product based on the exfoliator used to understand consumer behaviour and their interest in microbeads in cosmetics. Respondents had multiple response options, but also had the option for additional explanations.

The average daily emission of microbeads (DEM, mg per person per day) from body and facial scrubs to the sewerage system was expressed in mg per person per day and was based on the survey data as well as information regarding the content of polyethylene microbeads in different cosmetics from the first part of the present study (Eqn (1)):

$$DEM = \frac{f \cdot V \cdot c}{t} \quad (1)$$

where f is frequency of application (per year), V is volume of personal care product per application (mL), c is the average concentration of polyethylene microbeads in the respective body or facial scrubs (mg/mL) and t is the number of days per year. The DEM has been calculated separately for body and facial scrubs.

The average concentration of polyethylene microbeads in the effluent from the WWTP (CPME) (mg/m³) was calculated according to Eqn (2):

$$CPME = \frac{DEM \cdot n \cdot e}{Q_w} \quad (2)$$

where, DEM is a daily emission of microbeads (mg per person per day) to the sewerage system, n is number of inhabitants in the city

of Ljubljana, e is the portion of microbeads released from the WWTP (from 0 to 1) and Q_w (m^3/d) is the daily flow rate of wastewater. Microbeads concentration in surface water (MCSW) (mg/m^3) was calculated as a mass balance between input of microbeads with the discharged wastewater and the dilution of the wastewater in the Ljubljanica River (Eqn (3)):

$$MCSW = \frac{Q_w \cdot CPME}{Q_{w+r}} \quad (3)$$

where, Q_w is a flow rate of wastewater (m^3/d), $CPME$ is estimated concentration of polyethylene microbeads in the WWTP effluent (mg/m^3) and Q_{w+r} is a total flow rate of wastewater and river water (m^3/d). The details of DEM, CPME and MCSW calculations are given in the Supplementary document.

3. Results and discussion

3.1. Analysis of microbeads from cosmetic products

Microbeads recovered from the facial (A, C) and body scrubs (B, D, E) were basically composed of white fine powder with some portion of red and blue particles in the products C and D, respectively. Only product E additionally contained fine brown particles, possibly husks and shell powder that were listed among its ingredients. The IR analysis (Fig. S1, Supplementary document) confirmed that the majority of microbeads in all products are made of polyethylene. Only in the case of product E, the analysis showed presence of a small amount of material that is not made of polyethylene, but we were unable to determine the composition of this material. The amount of microbeads varied across products but was on average higher in body relative to facial scrubs (Table 1). Average concentration of microbeads in body and facial scrubs was 4.82 g/100 mL and 0.74 g/100 mL, respectively.

The concentration of microbeads in cosmetic products identified in the present study is similar earlier studies from around the world: Gregory (1996) reported microbead concentrations between 0.19 and 6.91 g per 100 g in hand soaps and facial scrubs in New Zealand. Napper et al. (2015) found concentration in facial scrubs in the UK from a few grams up to 10 g per 100 mL, while in the USA facial scrubs contained mainly between 8 and 10 g microbeads per 100 mL (Chang, 2015). Although product A

contained the lowest concentration of microbeads it had the highest mean number of particles per mg which can be explained by the rather small average particle size. On the other hand, product D had the highest mean particles size and consequently a lower number of particles at a comparable concentration as product A. It means that from 100 mL of product A, about 1,305,360 particles can be released while from product D these numbers might be as low as 543,750. It is therefore evident, that the mass of microbeads in cosmetic products is not necessarily linked to the number of microbeads possibly entering sewerage system and ultimately the freshwater ecosystem.

In our study, all microbeads were of rather irregular shape (Fig. 1). Similar observations are, for instance, reported by Napper et al. (2015). The irregular, but typical microbeads shape can be attributed to the process of microbeads production. They are produced by ultrafine grinding in mills (Wirth and Korte, 2009) leading to fragments and shredded pieces. In addition, some manufacturers add spherical particles to enhance visual attraction for consumers such as coloured spheres that were found in product C. Microbeads from product D were more rounded in comparison to the other products and microbeads from product E were covered by fine dust, possibly by shell powder, which could, however, not be confirmed analytically. The structure of microbeads in the products A, B and C was smooth and non-porous. These samples also showed very low specific surface area (Table 1), while microbeads with a more structured surface, namely from the products D and E had several times higher specific surface areas.

A wide size range of microbeads was noticed in each product with sizes up to 1000 μm (Fig. 2). However and in accordance with Fendall and Sewell (2009), the majority of particles was smaller than 100 μm (mean sizes are given in Table 1). On average, body scrubs (i.e. products B, D and E) contained bigger particles while high number of smaller particles was found in facial scrubs (i.e., products A and C). This pattern might be explained by the potential wish of consumers for a smoother abrasive effect on face skin, which is more likely to be provided by smaller polyethylene particles.

3.2. Lab-scale wastewater treatment plant

During and after usage of scrubs, microbeads are washed into sewerage systems and travel to WWTPs. The removal efficiency of

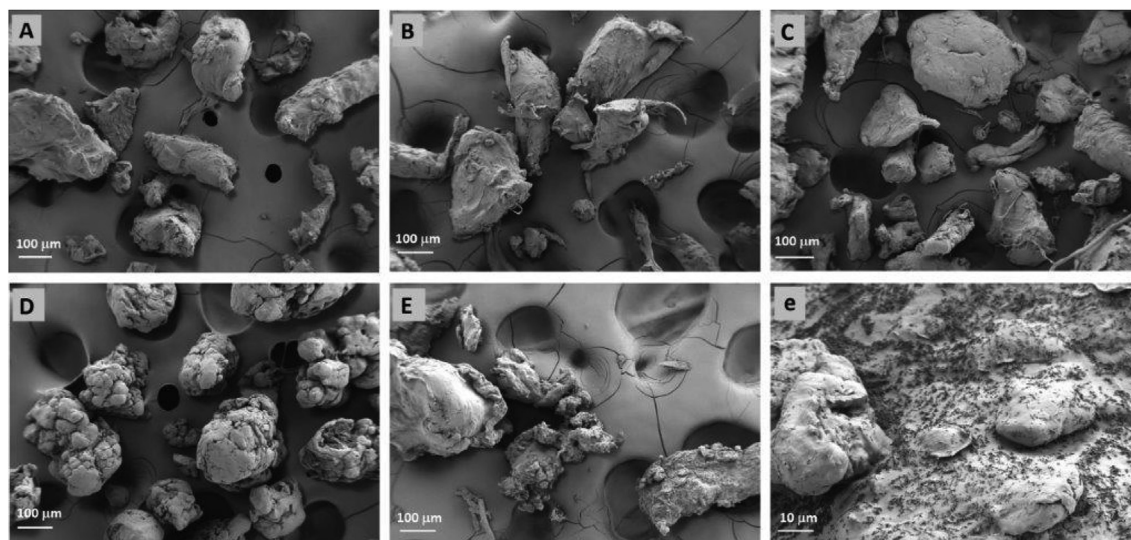


Fig. 1. SEM imaging of microbeads from two facial (A, C) and three body scrubs (B, D, E) with a ten-fold higher resolution of product E (e).

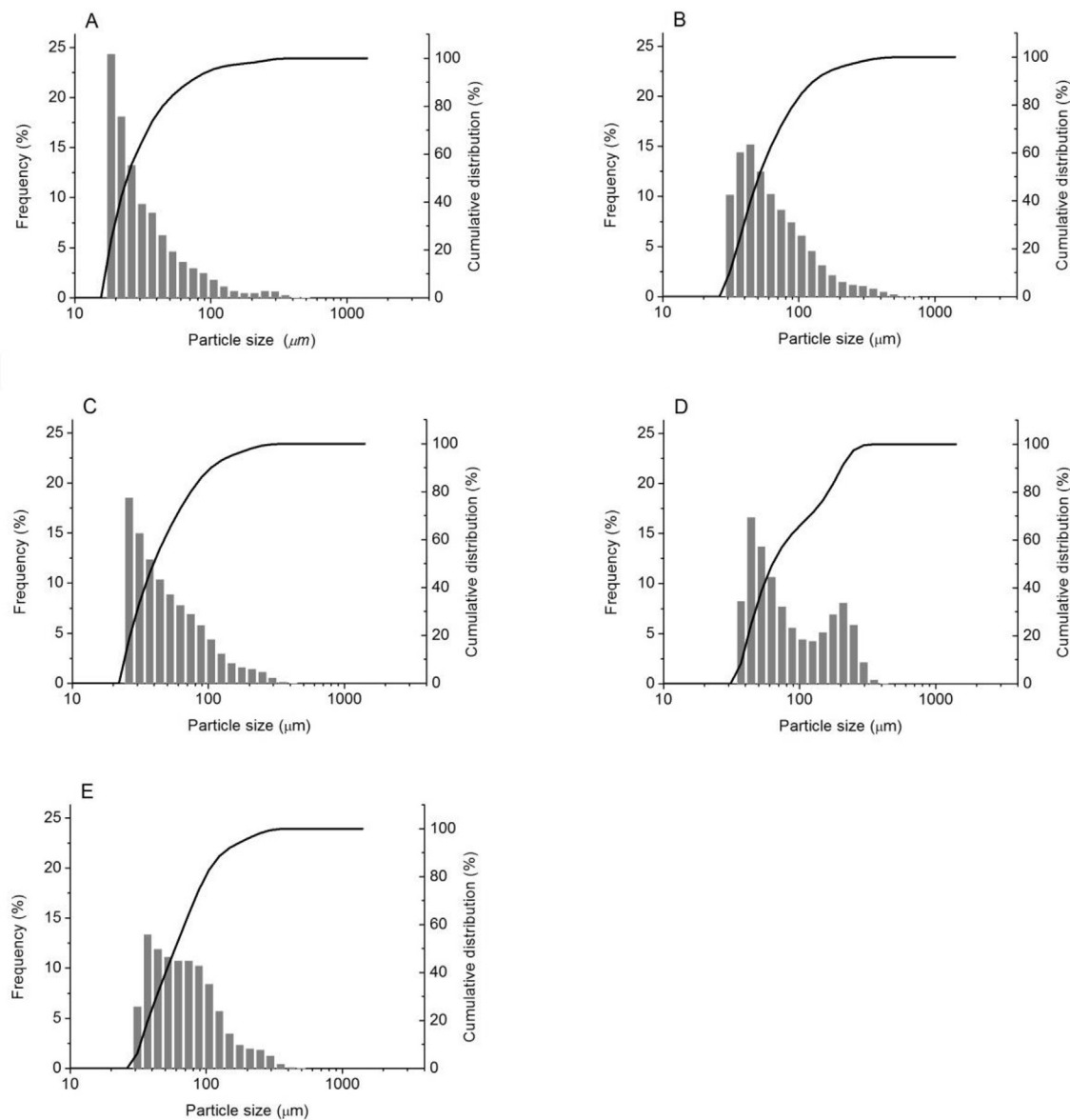


Fig. 2. Particle size distribution of microbeads from two facial (A, C) and three body scrubs (B, D, E).

the WWTPs for microplastics depends on involved treatment technology. The first treatment step in WWTPs is usually a primary treatment involving physical separation by screening and sedimentation, which is characterized by a microplastic removal efficiency of approximately 25% (see Chapter 2.2.). This suggests that the major load of microplastics is carried to the next step in the WWTP process. Indeed, Ziajahromi et al. (2017) indicated that polyethylene particles are among the most abundant microplastics in primary effluents. Moreover, these microbeads had the same shape and size range as microbeads from facial scrubs and toothpastes. These observations denote a high importance of personal care products as a source of microplastics. In contrast to the primary treatment, the secondary (i.e., biological) is considered more efficient and can reduce microplastic concentrations by 75% as reported by Talvitie et al. (2015). Similarly, Van Cauwenberghe et al. (2015) uncovered microplastic removal efficiencies during secondary treatment of approximately 44%, whereas others indicate values above 98% (e.g. Talvitie et al., 2017; Ziajahromi et al., 2017). These high efficiencies are, however, often linked to the availability

of tertiary treatments that include additional filtering (Talvitie et al., 2015; Carr et al., 2016), membranes (in this study reverse osmosis, Ziajahromi et al., 2017), dissolved air flotation or membrane bioreactor (Talvitie et al., 2017).

In the present study, we focused on a fate of polyethylene microbeads and not a complex mixture of microplastics during biological treatment. For this purpose microbeads from product B were used. In the first (MP1) and second (MP2) reactor 56% ($\pm 16\%$, $n = 6$) and 47% ($\pm 17\%$, $n = 6$) of the microbeads were removed (average removal efficiency was 52%). After each of the six treatment cycles, parts of microbeads were visibly captured in activated sludge settling to the bottom of the reactor. Although not measured, we assume that high affinity of polyethylene microbeads to negatively charged activated sludge flocs (Wilen, 1995) could be a consequence of a positive surface charge of polyethylene particles (Bhattacharya et al., 2010). Microbeads found after the treatment cycles were mainly floating on the water surface. Moreover, those microbeads recovered after the sixth cycle from treated wastewater of the reactor MP1 exhibited sizes above 60 μm with an average

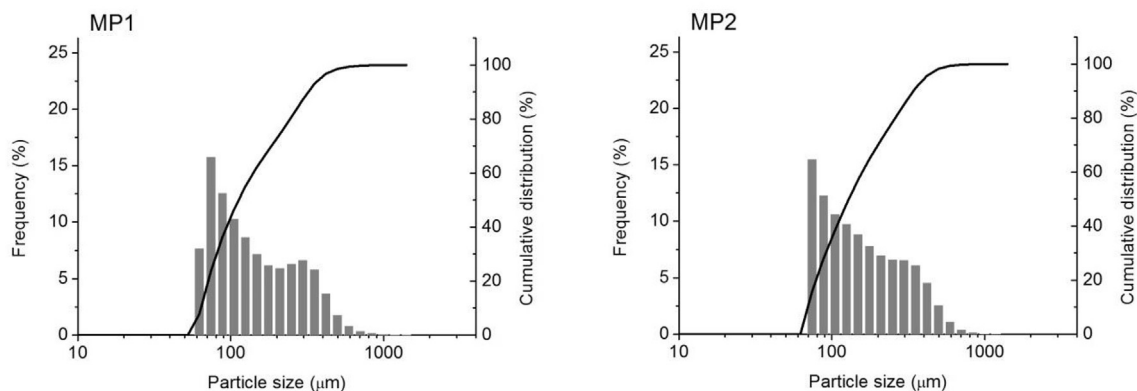


Fig. 3. Particle size distribution of microbeads recovered from the effluent of the reactors MP1 and MP2.

value of 159.3 μm (± 101.6 μm , $n = 3$), (10th and 90th percentile of the particle size distribution: 64.1 and 319.3 μm) (Fig. 3). Similar values were obtained from the reactor MP2: microbeads exhibited sizes from 70 μm with an average value of 172.7 μm (± 106.9 μm , $n = 3$), (10th and 90th percentile of the particle size distribution: 70.6 and 340.3 μm) (Fig. 3). These average microbead sizes are more than twice as high as the microbeads that were introduced into the reactors (Table 1, product B). This indicates that smaller particles are captured within the activated sludge while bigger particles remained in the water phase. The mean number of microbeads recovered from the reactors MP1 and MP2 were very similar; 119 and 106 particles per mg of microbeads, respectively (average particles per mg of microbeads was 112.5). It is about eight times less than the mean number of particles per mg added to the reactors (Table 1, Sample B) confirming a high retention efficiency (87%) of the biological wastewater treatment for the small fraction of the microbeads.

It is important to mention here, that the presence of microbeads did not affect efficiency of the SBR reactors. The removal of DOC in the control reactor was 93% ($\pm 7\%$, $n = 6$), while it was 93% ($\pm 8\%$, $n = 6$) and 95% ($\pm 7\%$, $n = 6$) in MP1 and MP2, respectively. Nitrification was also not inhibited and removal of NH_4^+ reached 98% ($\pm 1\%$, $n = 6$) in MP1, 97% ($\pm 1\%$, $n = 6$) in MP2, which is comparable to the control 96% ($\pm 3\%$, $n = 6$).

3.3. Estimation of daily emission of microbeads into the sewerage system and surface water exposure

For the estimation of daily emissions of microbeads from cosmetic products into sewage system 232 inhabitants of Ljubljana have been interviewed of which 178 were included in the study; participant with incomplete answers or incorrectly filled forms have been removed (additional information about the survey are given in the Supplementary document). Participants use on average 5.9 mL (± 9.5 mL, $n = 178$) of body scrubs and 3.5 mL (± 8.8 mL, $n = 178$) of facial scrubs per application. The frequency of application of body and facial scrubs is 17.2 and 25.7 times per year, respectively. If an average amount of microbeads of each scrub is taken into account the daily emission of microbeads (DEM, Eqn (1)) from body and facial scrubs in Ljubljana is 15.2 mg per person per day. Our result is about seven times higher than estimation in the U.S. (2.4 mg per person per day, market data, Gouin et al., 2011) and substantially lower than estimated for the U.K. (40.5–215 mg per person per day, market data, Napper et al., 2015). Nonetheless, the results of all these studies lie within the same range suggesting them as reasonable approximations based on the available information.

The results of our SBR study uncovered a retention of approximately 52% of the microbeads during secondary treatment in activated sludge. Combining these data with the published information on the removal of microplastics during primary treatment (see Chapter 2.2.) lead to an overall removal efficiency in a WWTP of approximately 77%. The concentration of polyethylene microbeads in effluents from the WWTP in the city of Ljubljana (288,179 inhabitants, RS, 2017) during average wastewater flow rates (72,567 m^3 , data obtained from the CWWTP Ljubljana) is estimated to be 13.9 mg/m^3 (DPME, Eqn (2)). This equals approximately 1 kg of microbeads released every day into the Ljubljana River (average flow rate of 5,184,000 m^3/d data from years 2006–2014, ARSO, 2017) resulting in a MCSW of 0.19 mg/m^3 (Eqn (3)). If the average number of particles released from a WWTP is taken into account (average of MP1 and MP2 reactor, 112.5 particles per mg of microbeads), about 112,500,000 particles can be released into the receiving river daily, resulting in microbeads concentration of 21 particles/ m^3 in Ljubljana River. Our calculation is in accordance with concentrations of microplastics found in the river Seine in France; concentrations from 3 to 100 microplastic (not limited to microbeads) particles/ m^3 (above 80 μm size) were lately reported (Dris et al., 2015).

3.4. Implications of the present study

Our results showed that treated wastewater can contain substantial amounts of microbeads, which may be substantially reduced by introducing a tertiary treatment step in WWTPs (Talvitie et al., 2017). At the same time, the microbeads released into receiving stream have particle sizes just above 60 μm . Since most of monitoring devices used a 333 μm mesh size as a cut off and can thus retain only particles above this size (Erkes-Medrano et al., 2015), the present study strongly suggested that the contribution of microbeads to the plastic litter pool (freshwater and marine) is substantially underestimated or even not detected. Furthermore, polyethylene microbeads showed a high affinity to activated sludge flocs which might suggest that these particles could preferentially accumulate in sediments, particulate organic matter and plants within the freshwater ecosystem. Hence, the transport dynamics of microplastics may be even more complicated than anticipated and certainly needs more attention.

These small and likely overlooked particles, however, potentially represent the most hazardous fraction of microplastics. This may be hypothesised as small particles may more easily be taken up by organisms opening doors for adverse effects and biotransfer (Cole et al., 2013; Jemec et al., 2016). Also the rather larger surface area to volume ratio increases the adsorption probability for co-

occurring contaminants supporting a co-transport of those contaminants into organisms during gut passage ultimately inducing negative effects (Koelmans et al., 2016).

4. Conclusion

The present study showed that high amounts of polyethylene microbeads are used in cosmetics with a rather small average particles size of around 100 μm . After application, 15.2 mg per person per day are released into the sewerage system. During wastewater treatment the majority of microbeads are captured in the activated sludge, but approximately 1 kg of microbeads was estimated to be released into receiving stream leading to predicted concentration of 21 particles/ m^3 in Ljubljana River. Considering that the present study suggests that microbeads are continuously released into freshwaters, the rather low concentrations estimated here might still be of concern for the environment given their high persistence in the environment. In addition, the likely underestimation of microbead exposure during current monitoring efforts suggests that environmental scientists have just scratched on the surface of this problem warranting further efforts to improve sampling and analytical methodologies. Those efforts should go hand in hand with a more detailed understanding on the transport dynamics in the water phase but also within aquatic food webs including associated negative impacts on the integrity of aquatic ecosystems.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.chemosphere.2017.08.131>.

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