Environ Monit Assess (2019) 191: 438
https://doi.org/10.1007/s10661-019-7574-1

Occurrence and risk assessment of microplastics from various toothpastes

Gul Sirin Ustabasi · Asli Baysal

Received: 1 April 2019 / Revised: 20 May 2019 / Accepted: 27 May 2019 / Published online: 15 June 2019

Abstract Microplastics have become a major environmental issue; their release from various products affects the aquatic environment. Personal care products such as toothpastes are recently being considered as a significant source of microplastics released to the aquatic environment. This study aims to assess the presence of microplastics found in toothpastes that are available in the drugstores and markets in Istanbul, Turkey. A total of 20 samples were tested. Following the extraction procedure, obtained particles were quantified and then characterized by microscopic evaluation and surface chemistry analysis. Twenty percent of the samples were found to contain microplastics in the structure of polyethylene at concentrations varying between 0.4 and 1%. In order to evaluate the release to environment, a risk assessment was conducted and yearly microplastic emission caused by toothpaste consumption was calculated based on the results.

Keywords Microplastics · Toothpaste · Risk assessment · Environment · Sea water

Introduction

Microplastics (MPs) are plastics with a size smaller than 5 mm (Jiang 2018). They can be either manufactured to have the certain size (primary MPs) or fragmented from larger plastics (secondary MPs). Most common types are polypropylene, polyethylene terephthalate, polymethyl methacrylate, nylon, and polyethylene (Lei et al. 2017). Their abundance and ubiquity are environmental concerns for the aquatic environment. Some milestones in the detection of MPs in various environmental samples are shown in Table 1. There are various studies indicating the existence of MPs found in aquatic systems and related components, such as lakes of India (Sruthy and Ramasamy 2017) and surface waters of China (Wang et al. 2017) and the USA (Eriksen et al. 2013). Additionally, Martin et al. (2017) found that MPs were present in the surface water and sediments (not deeper than 3.5 cm) in Irish continental shelf. Finally, MPs were even found in mineral water (Schymanski et al. 2018; Ossman et al. 2018) and table salts (Gundogdu 2018).

In order to evaluate their presence in the environment, their sources must be revealed. Among the main sources of MPs in the environment, one can find personal care and cosmetic products. Many personal care and cosmetic products, such as toothpastes, soaps, and gels, include MPs to enhance scrubbing and to strengthen their cleansing or exfoliating functions. It is estimated that 4360 tons of MPs are used in personal care and cosmetic products as additive agents each year in European Union countries (Lei et al. 2017; Anderson et al. 2016). The occurrence of MPs in cosmetic products has also been frequently reported in the last decade (Fendall and Sewell 2009; Lei et al. 2017; Napper et al. 2015; Godoy et al. 2019; Leslie 2014; Cheung and Fok 2016).

G. S. Ustabasi · A. Baysal (✉)
Health Services Vocational School of Higher Education, T.C. Istanbul Aydin University, Sefakoy Kucukcekmece, 34295 Istanbul, Turkey
e-mail: aslibaysal@aydin.edu.tr

Table 1 Chronological list regarding the occurrence of MPs in various sample types

Year	Sample type	Concentration	Observations	Reference
1972	Seawater	290 g/km^2	Brittle, pellet form	Carpenter and Smith Jr. (1972)
1974	Ocean surface water	–	White opaque and translucent pieces with various shapes	Colton et al. (1974)
1996	Hand cleaners and facial scrub	0.19–6.91 g/100 g product	Rough, shredded surface, white	Gregory (1996)
2006	Beach sediments and seawater	0–4 particles per sample replicate	Spherical	Ng and Obbard (2006)
2009	Facial cleansers	–	Various colors with granular and irregular shape	Fendall and Sewell (2009)
2011	Marine sediments, sewage and washing machine effluents	8–124 fibers per L sediment	Mostly synthetic fibers of textile	Browne et al. (2011)
2013	Surface waters of lake	43000 $particle/km^2$	Various colors in fragment, film, foam, and pellet form	Eriksen et al. (2013)
2014	Sub-surface seawater	8–9200 $particle/m^3$	Mostly fibers	Desforges et al. (2014)
2016	Sediments collected from beaches	In summer: 12–1300 and in winter: 3–743 $particle/m^2$	Various colors and shapes	de Carvalho and Baptista Neto (2016)
2017	Facial cleansers and shower gels	10.57–41.27 mg/g product	Various colors with granular and irregular shape	Lei et al. (2017)
2017	Sediments and bottom waters	7.67 MPs per station	Mostly fibers, and fragments	Martin et al. (2017)
2018	Table salts	8–102 item/kg	In the form of fibers, fragments, and film	Gundogdu (2018)
2018	Mineral water	2649–6292 MPs/L	Smaller than 5 μm	Ossman et al. (2018)
2018	Mineral water	11–118 particle/L	Smaller than 20 μm	Schymanski et al. (2018)
2019	Facial and body scrubs, exfoliating bath gels	0.99–7.80%	Various colors with spherical and irregular shapes	Godoy et al. (2019)

It is known that MPs can be added to the formulation to enhance whitening properties and act as smoothing and/or polishing agent (Leslie 2014; Vaz et al. 2018); however, examination of MPs in toothpastes was limited in the literature (Praveena et al. 2018; Hintersteiner et al. 2015).

In this study, we aimed to investigate the presence of MPs in commercially available toothpastes sold in Istanbul, Turkey. For this purpose, toothpastes of various brands were extracted, quantified, and characterized. Based on the data, risk assessment was conducted regarding their daily usage and comprised amounts. To our knowledge, it is the first study to reveal the contribution of toothpastes to MP contamination in Turkey.

Materials and method

Chemicals and instrumentation

Filter papers with particle retention of 4–7 μm were purchased from Macherey-Nagel (Germany). No plastic lab-ware was equipped during extraction, all the flasks and funnels were made of glass, and spatulas used for scraping the dry matters were made of steel.

To characterize and image the MPs, Fourier transform infrared spectrometer (FTIR; Bruker VERTEX 70ATR, Karlsruhe, Germany) and a light microscope with an attached AxioCam ICc 1 camera (Zeiss Scope.A1, Jena, Germany) were used.

Table 2 List of samples given with their assertions

Sample	Assertion
1	Protection against acid erosion and decay for kids
2	Protection against sensitivity
3	Whitening with calcium and fluoride
4	Cleans stains, freshens breath for smokers
5	Whitening with micro-cleansing crystals
6	Whitening and shining
7	12-hour protection
8	Toothpaste and mouthwash
9	Protects against sugar
10	Relief and sensitivity protection
11	Whitening
12	Whitening
13	Natural whitening and protection against decay
14	Cleans the stains caused by smoking
15	Cleans stains and whitens teeth
16	Whitening, refreshing, and protection against decay
17	Protection against sensitivity
18	Crystal mint freshness
19	100% herbal, complete care
20	Bio-active, whitening toothpaste

Sampling and extraction of MPs

All samples available in different prices and brands were purchased from cosmetic stores and markets in Istanbul, Turkey, and list of samples is given in Table 2.

Applied extraction method is summarized in Fig. 1. Ten grams of toothpaste was weighed and mixed with 500 mL deionized water via magnetic stirring while heating. After the entire sample was dissolved, filtration began immediately. Samples were left to be filtrated on their own with the help of gravity. After the filtration was completed, filtrates were dried in an oven at 50 °C for 7–8 h. Once they were cooled down to room temperature, they were weighed and the percentage of undissolved solids in the sample was calculated. Once the filtration was complete, obtained solids were mixed with 50 mL distilled water and present MPs in the structure of polyethylene were expected to float due to its density being lower than water. The mixtures were left to settle for 2 h. After the particles were settled, the liquid part was decanted to flasks made of glass to obtain floated particles and then dried at 75 °C. Weights of the dried solids were recorded. Finally, they were identified by FTIR.

Risk assessment

Risk assessment was carried out to find out the contribution of toothpaste to MP pollution in Istanbul, Turkey. For this purpose, findings of this study were utilized to determine the maximum, minimum, and mean values of emission. In order to find out the amount of toothpaste required for a single use, samples from all specimens were weighed and an average value was obtained. For the calculation of the maximum value, it was assumed that toothpastes were used by everyone twice a day, as suggested, and for the minimum value, toothpastes were assumed to be used once a day. The estimated MP emission values are calculated with the following formula, modified from the work of Cheung and Fok (2016) and Praveena et al. (2018):

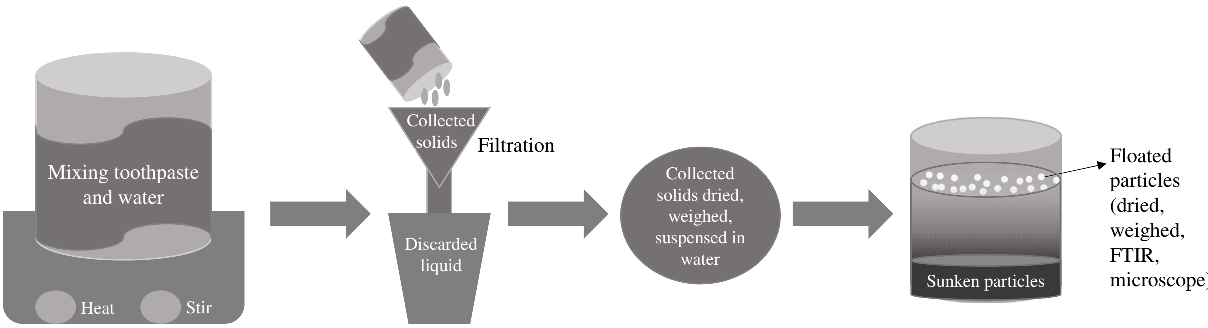

Fig. 1 Visual summary of the methods applied for MPs extraction from the toothpaste samples

$$YME = DU \times POP_{ist} \times TP_{use} \times OR \times MP \times N_{days}$$

where

YME yearly MP emission

DU number of daily usage

POP_{ist} total population of Istanbul

TP_{use} toothpaste amount required for single use

OR occurrence ratio of MPs in total number of samples

MP MP percentage in samples

N_{days} number of days in a year

Results and discussion

In order to detect and quantify the MPs in toothpastes, at first, obtained solid forms after the extraction were calculated as total undissolved solids, floated particles, and their percentage in product. These results are shown in Table 3.

To characterize the solid forms obtained from toothpaste extraction, FTIR and microscopic analysis were applied. It is known that MPs can be characterized by the presence of polypropylene, polyethylene terephthalate, polymethyl methacrylate, nylon, or polyethylene (Lei et al. 2017). In the FTIR analysis, polyethylene gives characteristic signals at 2915, 2848, 1460, and 716 cm^{-1} (Cheung and Fok 2016; Jung et al. 2018). According to the FTIR spectrums, four samples contained polyethylene which are 2, 3, 12, and 20 (Fig. 2). The samples including MPs by the occurrence of polyethylene had 0.039, 0.10, 0.043, and 0.064 g of floated particles in 10 g of samples 2, 3, 12, and 20, respectively. As shown in Table 3, solids obtained from other samples were also characterized by FTIR and contained various types of additives such as mica, $CaCO_3$, and $Ca_3(PO_4)_2$. These Ca-containing additives are generally added to the formula for polishing and whitening purposes (Carretero and Pozo 2010), whereas mica is often used for enhanced scrubbing effect. Solids obtained from samples 5, 6, 10, and 14 could not be identified.

In addition to FTIR characterization, the samples which included polyethylene were viewed under a microscope. Analysis by microscopy reveals that they have mostly irregular shapes with opaque appearance, as can be seen in Fig. 3. On the other hand, samples 3, 12, and 20 had also transparent/colorless objects that have square-like shapes. The images showed that particle sizes are smaller than 20 μm. Overall, particle sizes can be measured between 4 and 20 μm, because filter papers used for extraction were able to collect particles larger than 4 μm. Parallel to our study, Praveena et al. (2018) examined particle sizes of MPs in toothpaste and found the size in range of 3–145 μm.

To understand the contribution of a single source to MP contamination, occurrence of MPs should be determined in the representative samples of that source and emission rate should be calculated considering the population and usage frequency. In our study, we found that 4 out of 20 samples contained polyethylene as MPs and their concentrations varied between 0.4 and 1.0%. Furthermore, Hintersteiner et al. (2015) tested only one sample of toothpaste and found that 0.17% of the sample contained polyethylene-type MPs. In the study of Praveena et al. (2018), low-density polyethylene was found in one of the most popular toothpaste brands in Malaysia with less than 7% concentration. Brate et al. (2018) extracted approximately 100 mg of polyethylene from a 100-mL tube of a popular toothpaste brand with 50 μm size. Contrarily, Lei et al. (2017) reported that none of the toothpastes in their samples contained MPs.

To examine the risk of MPs release from toothpaste, we conducted an assessment using the data obtained in this study and consumption estimations for the residents

Table 3 Characterization and percentages of the particles obtained from toothpaste samples. *NI*, not identified; *PE*, polyethylene

Sample	Total undissolved solids, g	Floated particles, g	Weight in product, %	Composition
1	2.52	0.125	1.2	Mica
2	1.45	0.039	0.4	PE
3	4.02	0.105	1.0	PE
4	3.53	0.198	2.0	$CaCO_3$
5	2.68	0.043	0.4	NI
6	1.52	0.050	0.5	NI
7	3.66	0.057	0.6	$Ca_3(PO_4)_2$
8	1.21	0.074	0.7	Mica
9	1.95	0.029	0.3	$Ca_3(PO_4)_2$
10	2.10	0.075	0.8	NI
11	4.17	0.159	1.6	$CaCO_3$
12	0.39	0.043	0.4	PE
13	1.91	0.210	2.1	$Ca_3(PO_4)_2$
14	1.79	0.061	0.6	NI
15	2.62	0.140	1.4	$Ca_3(PO_4)_2$
16	2.26	0.147	1.5	$Ca_3(PO_4)_2$
17	1.53	0.150	1.5	Mica
18	2.34	0.025	0.2	Mica
19	1.94	0.061	0.6	Mica
20	3.66	0.064	0.6	PE

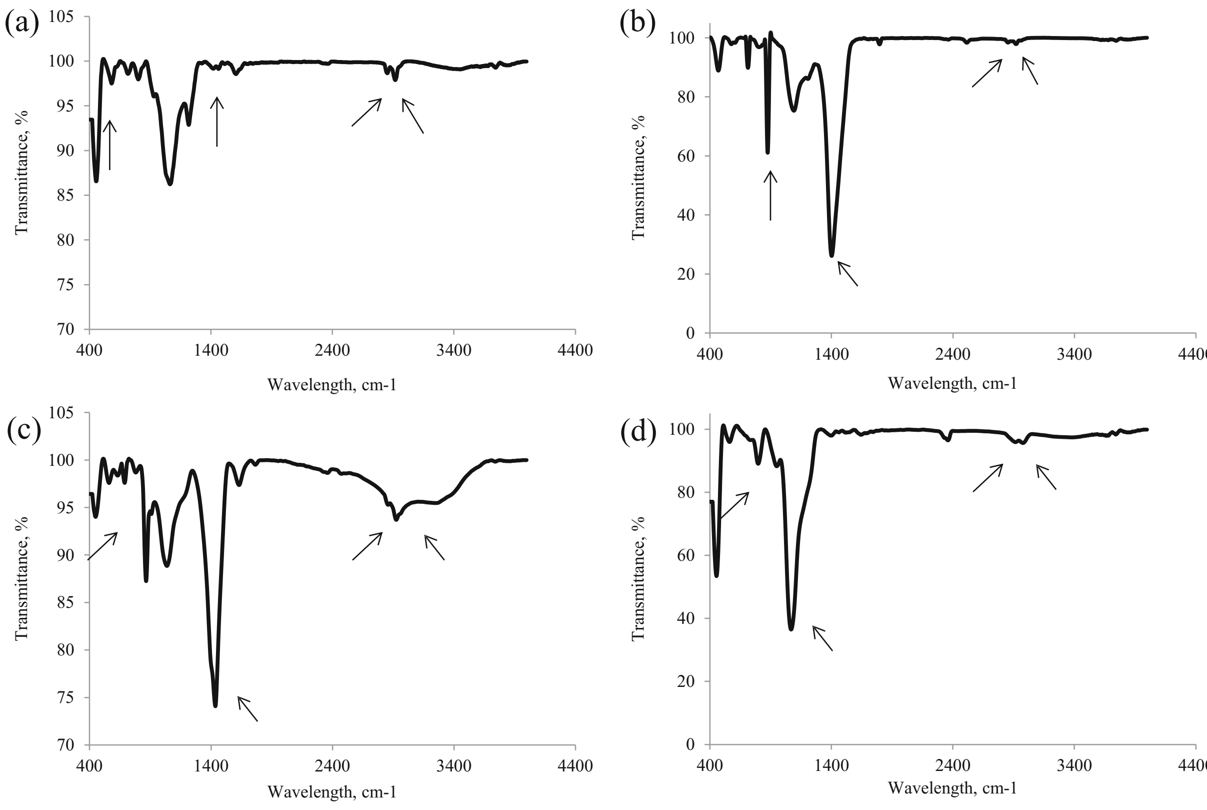

Fig. 2 FTIR results of particles extracted from samples **a** 2, **b** 3, **c** 12, and **d** 20. Characteristic peaks of PE are marked with grey arrows

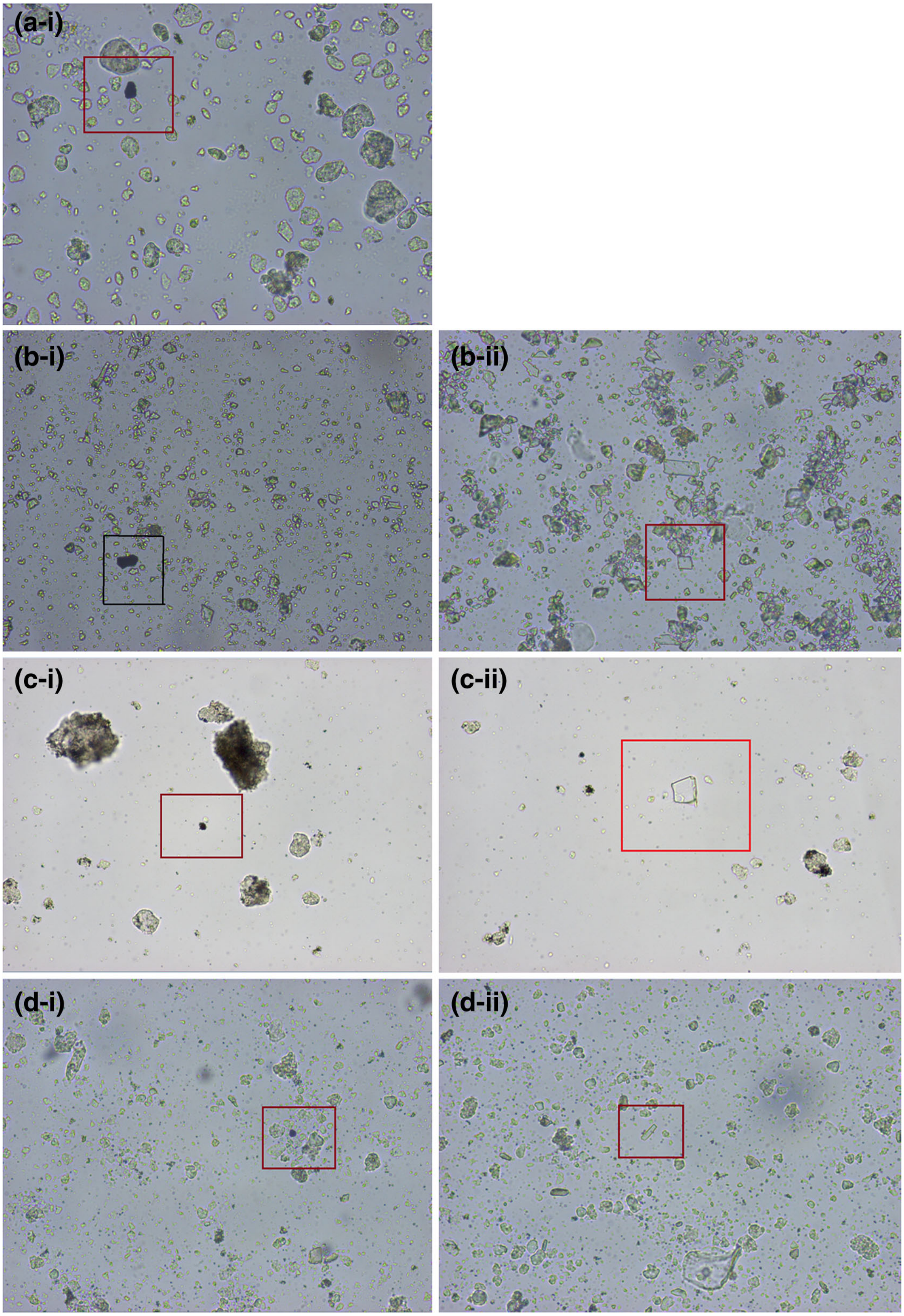

Fig. 3 Microscopic views of extracted MPs in samples of **a** 2, **b** 3, **c** 12, and **d** 20 under × 40 magnification. "i" represents opaque particles and "ii" represents transparent particles. Scale bar (black line): 20 μm

Table 4 Yearly emissions of MPs caused by the MPs in toothpaste for the city of Istanbul, Turkey

Parameters	Max. value	Min. value	Mean value	Reference
DU	2	1	1.5	Assumed
POP_{ist}	15,067.724	15,067.724	15,067.724	Turkish Statistical Institute (TSI) 2018
TP_{use}, g	1.40	0.50	0.88	This study
OC	0.2	0.2	0.2	This study
MP, %	1.0	0.4	0.6	This study
YME for Istanbul, g	3 billion	220 million	871 million	

of Istanbul. There are a limited number of studies in the literature evaluating the risks of contamination from different sources (Cheung and Fok 2016; Praveena et al. 2018). Cheung and Fok (2016) conducted a risk assessment and estimated that yearly, 342.2 billion microbeads were released to the environment from solely facial scrubs in Hong Kong. Praveena et al. (2018) reported the total emission of microbeads from personal care products (five facial cleansers and five toothpastes) as 0.199 trillion per year. However, there were some limitations such as examining mixed sample types and restrictions on age and population. Thus, to make an accurate estimation, it is important to calculate the contribution of a single source to environmental contamination. For this purpose, we conducted a modified risk assessment based on the works of Cheung and Fok (2016) and Praveena et al. (2018). As can be seen in Table 4, the risk assessment showed that emission of MPs from toothpaste usage in Istanbul can cause the release of yearly 220 million–3 billion g (average 871 million g) of MPs by the joining domestic wastewater stream and draining to seas or rivers. However, information on the escape rate of MPs from wastewater treatment plants of Istanbul Water and Sewerage Administration (ISKI) is unknown; escape rates are mainly determined by the technology used in treatment plants, and it was reported that current wastewater treatment plants have the inability to capture MPs efficiently (Roex et al. 2013; Kay et al. 2018).

Conclusion

In this study, 20 different toothpaste samples were examined to detect and quantify MPs. Four samples were found to have polyethylene in varying amounts. Considering the data obtained from the study, yearly 871 million g of MPs on average is estimated to be emitted from toothpastes in Istanbul, Turkey. To our knowledge, this is the first study to thoroughly investigate toothpastes for MPs and calculate the yearly emissions in Istanbul. The concerns for possible threats of MP contamination in aquatic life continue to rise due to unavoidable usage and accumulation of those particles in the environment with increasing population. Further studies are needed to understand the fate and impact of MPs in aquatic environment.

References

Anderson, A. G., Grose, J., Pahl, S., Thompson, R. C., & Wyles, K. J. (2016). Microplastics in personal care products: Exploring perceptions of environmentalists, beauticians and students. *Marine Pollution Bulletin, 113*, 454–460.

Brate, I. L. N., Blazquez, M., Brooks, S. J., & Thomas, K. V. (2018). Weathering impacts the uptake of polyethylene microparticles from toothpaste in Mediterranean mussels (M. galloprovincialis). *Science of the Total Environment, 626*, 1310–1318.

Browne, M. A., Crump, P., Niven, S. J., Teuten, E., Tonkin, A., Galloway, T., & Thompson, R. (2011). Accumulation of microplastics on shorelines worldwide: Sources and sinks. *Environmental Science and Technology, 45*, 9175–9179.

Carpenter, E. J., & Smith, K. L., Jr. (1972). Plastics on the Sargasso Sea surface. *Science, 175*(4027), 1240–1241.

Carretero, M. I., & Pozo, M. (2010). Clay and non-clay minerals in the pharmaceutical and cosmetic industries Part II. Active ingredients. *Applied Clay Science, 47*(3–4), 171–181.

Cheung, P. K., & Fok, L. (2016). Evidence of microbeads from personal care product contaminating the sea. *Marine Pollution Bulletin, 109*, 582–585.

Colton, J. B., Knapp, F. D., & Burns, B. R. (1974). Plastic particles in surface water of Northwestern Atlantic. *Science, 185*, 491–497.

de Carvalho, D. G., & Baptista Neto, J. A. (2016). Microplastic pollution of the beaches of Guanabara Bay, Southeast Brazil. *Ocean & Coastal Management, 128*, 10–17.

Desforges, J. P., Galbraith, M., Dangerfield, N., & Ross, P. S. (2014). Widespread distribution of microplastics in subsurface seawater in the NE Pacific Ocean. *Marine Pollution Bulletin, 15*(79), 94–99.

Eriksen, M., Mason, S., Wilson, S., Box, C., Zellers, A., Edwards, W., Farley, H., & Amato, S. (2013). Microplastic pollution in the surface waters of the Laurentian Great Lakes. *Marine Pollution Bulletin, 77*(1–2), 177–182.

Fendall, L. S., & Sewell, M. A. (2009). Contributing to marine pollution by washing your face: Microplastics in facial cleansers. *Marine Pollution Bulletin, 58*, 1225–1228.

Godoy, V., Martin-Lara, M. A., Calero, M., & Blazquez, G. (2019). Physical-chemical characterization of microplastics present in some exfoliating products from Spain. *Marine Pollution Bulletin, 139*, 91–99.

Gregory, M. R. (1996). Plastic 'scrubbers' in hand cleansers: A further (and minor) source for marine pollution identified. *Marine Pollution Bulletin, 32*, 867–871.

Gundogdu, S. (2018). Contamination of table salts from Turkey with microplastics. *Food Additives and Contaminants, 35*(5), 1006–1014.

Hintersteiner, I., Himmelsbach, M., & Buchberger, W. W. (2015). Characterization and quantitation of polyolefin microplastics in personal-care products using high-temperature gel-permeation chromatography. *Analytical and Bioanalytical Chemistry, 407*, 1253–1259.

Jiang, J. Q. (2018). Occurrence of microplastics and its pollution in the environment: A review. *Sustainable Production and Consumption, 13*, 16–23.

Jung, M. R., Horgen, F. D., Orski, S. V., Rodriguez, V., et al. (2018). Validation of ATR FT-IR to identify polymers of plastic marine debris, including those ingested by marine organisms. *Marine Pollution Bulletin, 127*, 704–716.

Kay, P., Hiscoe, R., Moberley, I., Bajic, L., & McKenna, N. (2018). Wastewater treatment plants as a source of microplastics in river catchments. *Environmental Science and Pollution Research, 25*, 20264–20267.

Lei, K., Qiao, F., Liu, Q., Wei, Z., Qi, H., Cui, S., Deng, Y., & An, L. (2017). Microplastics releasing from personal care and cosmetic products in China. *Marine Pollution, Bulletin, 123*, 122–126.

Leslie, H. A. (2014). *Review of microplastics in cosmetics*. Amsterdam: Institute for Environmental Studies VU University.

Martin, J., Lusher, A., Thompson, R. C., & Morley, A. (2017). The deposition and accumulation of microplastics in marine sediments and bottom water from the Irish Continental Shelf. *Scientific Reports, 7*, 10772–10781.

Napper, I. E., Bakir, A., Rowland, S. J., & Thompson, R. C. (2015). Characterisation, quantity and sorptive properties of microplastics extracted from cosmetics. *Marine Pollution Bulletin, 99*, 178–185.

Ng, K. L., & Obbard, J. P. (2006). Prevalence of microplastics in Singapore's coastal marine environment. *Marine Pollution Bulletin, 52*(7), 761–767.

Ossman, B. E., Sarau, G., Holtmannspoetter, H., Pischetsrieder, M., Christiansen, S. H., & Dicke, W. (2018). Small-sized microplastics and pigmented particles in bottled mineral water. *Water Research, 141*, 307–316.

Praveena, S. M., Shaifuddin, S. N. M., & Akizuki, S. (2018). Exploration of microplastics from personal care and cosmetic products and its estimated emissions to marine environment: An evidence from Malaysia. *Marine Pollution Bulletin, 136*, 135–140.

Roex, E., Vethaak, D., Leslie, H., & de Kreuk, M. (2013). *Potential risk of microplastics in the fresh water environment*. Amersfoort: STOWA.

Schymanski, D., Goldbeck, C., Humpf, H. U., & Fuerst, P. (2018). Analysis of microplastics in water by micro-Raman spectroscopy: Release of plastic particles from different packaging into mineral water. *Water Research, 129*, 154–162.

Sruthy, S., & Ramasamy, E. V. (2017). Microplastic pollution in Vembanad Lake, Kerala, India: The first report of microplastics in lake and estuarine sediments in India. *Environmental Pollution, 222*, 315–322.

Turkish Statistical Institute (TSI). (2018). Population of provinces by years. http://tuik.gov.tr/UstMenu.do?metod=temelist Last access date: 11 March 2019.

Vaz, V. T., Jubilato, D. P., Oliveira, M. R. M., Bortolatto, J. F., Floros, M. C., Dantas, A. A. R., & Oliveira Junior, O. B. (2018). Whitening toothpaste containing activated charcoal, blue covarine, hydrogen peroxide or microbeads: which one is more effective? *Journal of Applied Oral Science, 27*, e20180051.

Wang, W., Ndungu, A. W., Li, Z., & Wang, J. (2017). Microplastics pollution in inland freshwaters of China: A case study in urban surface waters of Wuhan, China. *Science of the Total Environment, 575*, 1369–1374.