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ORIGINAL PAPER

Microplastics and endocrine-disrupting chemicals released from disposable hot beverage cups and from teabags, and their evaluation in terms of human health safety*

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Abstract

Microplastics (MPs) are pollutants that contaminate resources such as water, air, and soil. Humans can be exposed to microplastics through inhalation, digestion, or dermal contact. The use of disposable paper and plastic cups, as well as teabags, is widespread. Cups are usually made of plastic, and teabags are generally made of half-synthetic cellulose. This study aims to draw attention to their effects on human health by analyzing MPs due to contact with plastic paper cups and teabags with a hot aqueous solution (50°C, 70°C, and 90°C). This study used scanning electron microscopy (SEM), high-performance liquid chromatography, and mass spectrometry (LC-MS/MS), and the results were confirmed by Fourier transform infrared spectroscopy (FTIR). The number of MP particles released from teabags increased to 4.59 x 10^4 particles cm^{-3}, while in paper cups and plastic cups, MP particle numbers elevated to 2.94 x 10^4 particles cm^{-3} and 2.11 x 10^4 particles cm^{-3}, respectively. Total phthalate compound concentrations determined in aqueous solution samples were 1.74-2.42 μg dm^{-3} in teabags, 1.49-2.40 μg dm^{-3} in paper cups, 1.20 - 1.28 μg dm^{-3} in plastic cups; Bisphenol A concentration ranges were determined as 0.015-0.045 μg dm^{-3} in plastic cups, 0.006-0.011 μg dm^{-3} in teabags and 0.005-0.006 μg dm^{-3} in paper cups. Consumption of tea or coffee that comes into contact with disposable plastics daily may increase the daily MP intake and cause health problems in the future.

Keywords: microplastics, health, paper cup, plastic cup, teabag

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INTRODUCTION

Nowadays, plastics are used in many areas, such as the manufacture of cosmetics (Juliano, Magrini 2017, Jacob et al. 2020), textiles, furniture (Abbasi et al. 2019), food packaging, disposable kitchen products, and in the pharmaceutical industry (Patel et al. 2009). For this reason, their production has been increasing daily for the last 30 years. This has led to plastics being found in every aspect of our daily activities. Plastic pollution has occurred due to the accumulation of plastic wastes in landfills and in the seas because of the insufficient recycling of plastic waste in a controlled and effective manner despite the increasing production and low biodegradation of plastics in the ecosystem.

Plastic waste with a diameter over 5 mm, called macroplastics, decomposes under the influence of environmental factors, such as temperature, sun rays, wind, and erosion, causing the formation of MPs and nanoplastics (Andrady 2011, Zettler et al. 2013). MPs are carbon-polymer-based compounds with sizes between 5 mm and 100 nm, while NPs include particles less than 100 nm in size (Thompson et al. 2004, Rios 2007). While some are called primary MPs, namely ones produced in micro-sizes are to be used during the production of consumables, such as paper and plastic cups, teabags, clothes, toothpaste, face wash products, and shower gels, which we frequently use in our daily life; those formed due to the breakdown or degradation of large plastics are called secondary MPs (Lei et al. 2017, Karbalaei et al. 2018).

Both primary and secondary MPs can be transported to distant regions with the help of air currents, water currents, and human-induced factors (Karbalaei et al. 2018). MPs mixed with air or food can be transmitted directly to humans. Those that blend with the soil can be transferred to humans by being included in the food chain because they access spring waters, sea waters, sea creatures, and plants (Bouwmeester et al. 2015).

Bottled potable water and seafood are essential in MP exposure (Jacob et al. 2020). The mean MP count in a drinking water bottle was 188±88 particles dm^{-3} (Schymanski et al. 2018). The same study showed that the plastics in bottled drinking water were polyethylene terephthalate (PET) and polypropylene (PP), confirming the information that MP was transferred from packages to beverages (Schymanski et al. 2018). In today's modern society, there is an increase in the use of single-use plastic beverage cups and takeaway food containers. In a study to determine the microplastic contents, elevated levels of microplastics were found, with up to a million microparticles/mL and submicron particles (Liu et al. 2022). In hot water, disposable plastic materials produce hazardous chemicals and microplastics. Approximately 25,000 micron-sized microplastic particles were found in one cup of hot water (100 cm^{-3}) in another investigation that identified and quantified the microplastic particles leaching into the liquid. Toxic heavy metals,

like Pb, Cr, and Cd, were found in addition to microplastic particles (Ranjan et al. 2021).

Although it is currently impossible to quantify the health hazards associated with microplastic exposure in people accurately, attempts are being undertaken in this direction. In a different study, some parameters were determined, including chronic daily intake (CDI) and lifetime intake (LTI), which reflect the possibility of microplastic exposure through ingesting food and beverages and MP consumption through the ingestion pathway. According to the findings (Joseph et al. 2023), CDI could accumulate up to 0.03 to 0.025 mg of microplastic kg^{-1} of body weight.

However, (PE), polypropylene (PP), polyethylene terephthalate (PET), polyvinyl chloride (PVC), and polystyrene (PS) account for more than 90% of the polymers used in the production of plastic food packaging and containers (Alojaly, Benyounis 2020).

During the production of plastics, some chemical additives (bisphenol A, phthalates) are used to increase their durability and reduce their biodegradation (Ferreira et al. 2019). MPs can pose a chemical and physical threat to human health due to both the additives they contain and their small size (Prata et al. 2020, Zhang, Xu 2022).

To determine the potential effects of MPs on human health, it is vital to consider how much MPs can be released into foodstuffs that come into contact with plastic containers and products that may contain plastic (Chen et al. 2023). Humans are exposed to MPs through inhalation, skin contact, and the digestive tract by foods and beverages. MPs are a permanent pollutant owing to their resilience towards environmental factors. Besides that, the deposition of MPs in living beings without changing their structure is shown to affect human health by causing oxidative stress, cytotoxicity, inflammation, obstruction, and toxicity toward lipid metabolism (Wright et al. 2013, Wang et al. 2016, Smith et al. 2018, Rahman et al. 2021).

In today's modern societies, people prefer disposable products to create time for themselves (Kayan, Küçük 2020). Among these products, disposable cups are widely used in many coffee and tea-selling places worldwide (Poortinga, Whitaker 2018). These cups are of two types: plastic and plastic coated paper cups (Chen et al. 2023). In paper cups, 90-95% of their weight is paper, the remaining mass, especially the inner surfaces that come into contact with the beverage, is mainly covered with a water repellent plastic layer made of polyethylene (Rogovina et al. 2013, Mitchell et al. 2014, Constant 2016, Arumugam et al. 2018).

Phthalates, antioxidants (phenolics, organophosphates), perfluoroalkyl (PFAs), bisphenol A (BPA), and other additives are used to give the desired properties (flexibility, durability, lightness, antimicrobial properties, water, and oil repellent properties) to plastics used in food packaging. (Lopez--Espinosa et al. 2007, Chang, Chen 2010, Trier et al. 2011, Fierens et al. 2012, Souza and Fernando 2016, Schaider et al. 2017, Hahladakis et al.

2018, Schultes et al. 2019, Xue et al. 2019). In addition, it has been shown that the contact of 1 teabag made of plastic or plastic-containing materials with hot water significantly increases MP release and releases an average of 11.6 billion MP and 3.1 billion nanoplastics to a cup of hot water (Hernandez et al. 2019, Li et al. 2020).

The chemicals known as endocrine disrupting chemicals (EDC) [bisphenols, phthalates, polychlorinated biphenyls] and their derivatives [bisphenol A, bisphenol S, polyvinyl chloride (PVC), dimethyl phthalate (DMP), dibutyl phthalate (DBP) and di(2ethylhexyl) phthalate (DEHP)…] are commonly found in the environment due to their abundant use in daily life (Ahn, Jeung 2023, Wang et al. 2023). When the data form the latest studies are examined, it is claimed that EDC exposure, especially in the utero, might be responsible for the uptick in the number of patients with chronic diseases, such as diabetes mellitus, obesity, hypertension, hormone-related cancers, and infertility in the last 2-3 decades (Soto, Sonnenschein 2010, Bonde et al. 2017).

Since disposable cups and teabags are used extensively worldwide and this use is usually associated with the use of hot water, it is crucial to investigate the transfer of MPs to such beverages. This study aims to examine the MP releases of plastic cups, paper cups, and teabags that may contain plastic, to the hot beverage they contact with, to determine whether there are EDC releases that these disposable products may include, to determine the amounts, if any, and to draw attention to their effects on human health. This study also aims to explain the polymer structures of released MPs in hot water solutions using ATR-FTIR analyses.

MATERIALS AND METHODS

Collection of plastic materials

Plastic cups, paper cups, and teabags were purchased from the local markets in Edirne /Türkiye; samples were brought as five items per each type of cup sold under the same brand from three different stores. A total of fifteen samples for plastic cups and an equal number of pieces for paper cups were used in the experiments. Samples for teabag experiments and analyses were acquired from the same store (as three packages each contain 20 teabag/pack) apart from cups, and fifteen randomly selected teabags were used in experiments. All samples were stored in a dark place until the experiments and analysis were conducted.

Sample preparation and analysis

Analysis of MPs in cups and teabags

The sample preparation procedure was applied to all plastic and paper cups taken as samples. Ultra-pure water (UPW), water with Milli-Q purity, was obtained from the ultra-purification system (Merck Millipore-Direct Q 3UV, Darmstadt, Germany) and used in all experiments.

The temperatures of the distilled water solutions in beakers with a volume of 1.0 dm^3 were increased to different temp. of 50°C, 70°C, and 90°C using hot-plate heaters; 200 mL portions were taken from these volumes and transferred to plastic and paper cups. For teabags, after emptying the contents, the bags were brought into glass beakers, and 200 cm^3 of water at different temperatures was added to them; three parallel samples were prepared for each temperature.

Water solutions in the glasses were mixed at a constant speed, avoiding contact with the glass baguettes and the vessel walls, for the same length of time (1 min), and then the solutions were left to cool down to room temperature. After keeping the teabags in the beaker for the specified time (5 min), they were taken out of the aqueous solution, and the contents of the cup were cooled to room temperature.

The resulting aqueous solutions were filtered using a nitrocellulose filter (pore size 0.45 μm, Sartorius Stedim Biotech, Gottingen, Germany), and MP particles released from cups and teabags were isolated from aqueous solutions. The filters were dried in a vacuum oven at 50°C (Pol-Eko-Aparatura SLN 115 STD, Wodzislaw Śląski, Poland), and MP particles were visualized under an optical microscope (Leica IC 80HD-M205C, Wetzlar, Germany) – Figure 1.

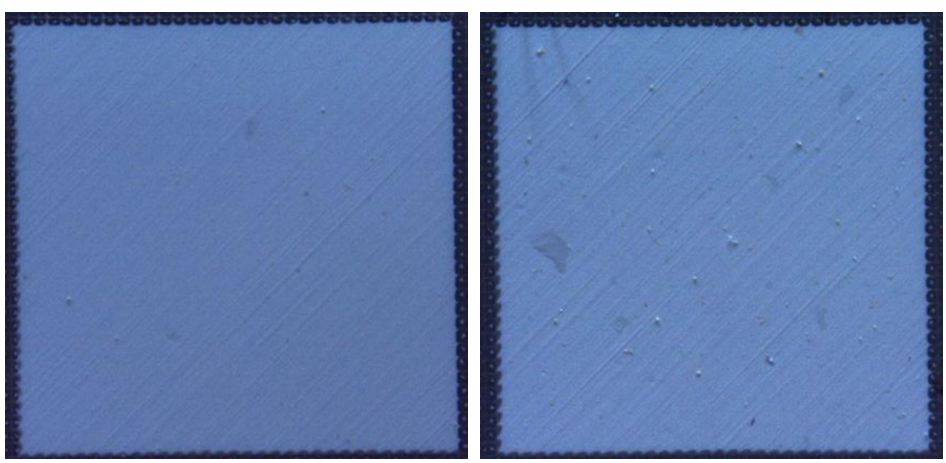

Fig. 1. Microplastic particles observed (released from plastic cup into 200 cm^3 at 50°C, left one is a blank sample) on nitrocellulose filter (1 cm^2 area) under the optic microscope (Microscope: MST5x-DCI, Camera: Leica IC80 HD Camera, Exposure: 85.4 ms, Gain: 12.0 x, Gamma: 0.8)

Before counting MP particles under a scanning electron microscope (SEM), 5.10^{-4} dm^3 (500 µL) of solution taken from the samples were transferred onto an aluminum coverslip, and the water was evaporated in a vacuum oven at 50°C, and SEM analyses (SEM-Zeiss EVO LS-10, Carl Zeiss NTS, Germany) was carried out. Images of MPs were performed at magnifications of 1000x -5000x- 10000x for 0.5-5 µm sized particles. Counting was completed by taking the average values of at least 3 replicate analyses for each sample.

The chemical structure of MPs was elucidated by the Attenuated Total Reflectance on a Fourier Transform Infrared spectrophotometer (Frontier, ATR-FTIR, Perkin Elmer, US). For this purpose, 200 cm^3 of sample solutions prepared at three different temperatures were taken and the water volume was evaporated to dryness. MPs that could be found in the residue were taken to the solvent phase with 1 mL of methanol. All of the volumes of methanol solution were transferred dropwise onto the device's ATR surface, while evaporation of the methanol solvent was accomplished by airflow after each drop.

The ATR-FTIR spectrum of the MPs deposited on the ATR surface was scanned at 4500-400 cm^{-1} frequencies. The spectra and peaks obtained from the samples were interpreted using the relevant studies in the literature, and the structures of MPs were defined.

EDCs (Endocrine Disrupting Compounds), sample preparation and analysis

Dimethyl phthalate, diethyl phthalate, dipropyl phthalate, diphenyl phthalate, diphenyl iso phthalate, dibenzyl phthalate, benzyl butyl phthalate, dibutyl phthalate, bis (4-methyl pentyl) phthalate, bis(2-ethyl hexyl) phthalate, di-n-octyl phthalate) and BPA (Bisphenol A) compounds (Merck KgaA, Darmstadt, Germany) were selected for LC-MS/MS analysis.

Hydrophilic-hydrophobic balanced cartridges (Oasis HLB 6 cc, 500 mg LP extraction cartridge, Massachusetts, USA) were used as solid phase components in the purification processes with solid phase extraction (SPE).

In the SPE process, the cartridges were conditioned with 10 cm^3 of methanol followed by 10 cm^3 of UPW water. 200 cm^3 of samples of water solutions prepared as previously described (Section 1.2.1) were passed through cartridges at optimized flow rates (2 cm^3 min^{-1}).

The absorbent mass was dried by passing airflow through the cartridges with an SPE vacuum manifold at room temperature. In the elution process, 2x5 cm^3 of methanol: dichloromethane (1:1, v/v) solvent mixture was passed through the cartridges (1 cm^3 min^{-1}). The collected eluates were kept under nitrogen flow in a block heater (Stuart SBH 130 D, Bibby Scientific, Staffordshire, UK), and the solvents were evaporated to dryness; the solvent exchange was completed with 1.0 cm^3 of methanol/ethyl acetate (1:1, v/v).

Solutions containing SPE-purified analytes were analyzed by LC-MS/MS (Agilent 6460 Triple Quad LS/MS, CA, USA). Chromatogram and mass spectra obtained from LC-MS/MS were processed using the software program (Mass Hunter Workstation Data Acquisition (Version B.10.0).

The parameters for LC-MS/MS analysis were optimized, and mass spectra, including m/z values for molecular ions and fragments, were defined. Recoveries (%) of analyte compounds, detection limits (LOD), and quantification limits (LOQ) for the analytical method were set (Tables 1-4).

Table 1

LC-MS/MS analysis parameters for quantification of analytes

Analyte compounds	M (m/z)	Fragment ions (m/z)	FV (V)	CE (V)	ESI+/-
Diphenyl iso phthalate	319.0	225.1; 114.8	130	10: 60	+
Diphenyl phthalate	319.0	77.1; 224.9	80	40: 8	+
Dibenzyl phthalate	347.0	91.0; 181.0	90	20: 4	+
Dipropyl phthalate	251.0	148.9; 190.9	70	10: 2	+
Benzyl butyl phthalate	313.0	149.1; 91.0	80	8: 36	+
Dimethyl phthalate	194.9	163.0; 77.1	70	6: 36	+
Dibutyl phthalate	279.0	148.9; 205.0	80	52: 4	+
Diethyl phthalate	223.0	149.0; 177.0	70	14: 2	+
Di-n-octyl phthalate	391.1	148.8; 121.0	100	12: 52	+
Bis(2-ethylhexyl) phthalate	391.1	279.0; 148.8	110	4: 24	+
Bis(4-methyl pentyl) phthalate	335.1	148.9; 84.8	100	20: 8	+
Bisphenol A	227	211.9; 132.9	80	12; 22	-

M – molecular ion, FV – fragmentation potential, CE – collision energy

Table 2

Chromatographic separation parameters

Phthalates	A) Ultra-pure water (UPW) + 0.1% formic acid B) Methanol + 0.1% formic acid
LC column	Zorbax SB-C8, 3.0 x 150 mm, 3.5 µm
Temperature	20°C
Injection volume	5 µL (5.10^{-6} dm^3)
Flow rate	0.6 cm^3 min^{-1}
BPA	A) UPW; 5 mM ammonium acetate B) Acetonitrile; 10 mM ammonium acetate (19:1 v/v)
LC column	Zorbax SB-C8, 3.0 x 150 mm, 3.5 µm
Temperature	30°C
Injection volume	10 µL (1.10^{-5} dm^3)
Flow rate	0.6 cm^3 min^{-1}

upper – phthalates, lower – BPA

Table 3

Gradient elution parameters used in the chromatographic development

Phthalates			BPA		
Time (min)	A (%)	B (%)	time (min)	A (%)	B (%)
0.00	98.0	2.0	0.00	98.0	2.0
2.00	98.0	2.0	2.00	98.0	2.0
4.00	5.0	95.0	4.00	0.0	100.0
8.00	5.0	95.0	8.00	0.0	100.0
8.10	98.0	2.0	8.10	98.0	2.0
13.00	98.0	2.0	12.00	98.0	2.0

Table 4

Method-based, recovery efficiency, detection limits, and quantification limits

Compounds	Recoveries (%) (n=5)	LOD (ng cm^{-3})	LOQ (ng cm^{3})
Diphenyl iso phthalate	86	0.120	0.360
Diphenyl phthalate	87	0.037	0.111
Dibenzyl phthalate	85	0.286	0.858
Dipropyl phthalate	86	0.455	1.366
Benzyl butyl phthalate	87	0.337	1.011
Dimethyl phthalate	86	1.244	3.731
Dibutyl phthalate	87	0.525	1.576
Diethyl phthalate	88	0.038	0.114
Di-n-octyl phthalate	87	0.089	0.267
Bis(2-ethylhexyl) phthalate	89	0.030	0.089
Bis(4-methyl pentyl) phthalate	92	0.050	0.150
Bisphenol A	88	0.271	0.812

RESULTS

Optical microscope and SEM analysis results

After the MP particles were released from beverage containers into pure water, solutions were examined under an optical microscope, and SEM analyses were completed with three replicate (n=3) samples (Figure 2).

The percentile relative standard deviation (%RSD) values in the counts were calculated in the range of 1.3%-3.9%. The highest numbers of MP particles released into water solutions were determined at 90°C (Figure 2 upper image). In the microplastic particle counts for paper cups, an average

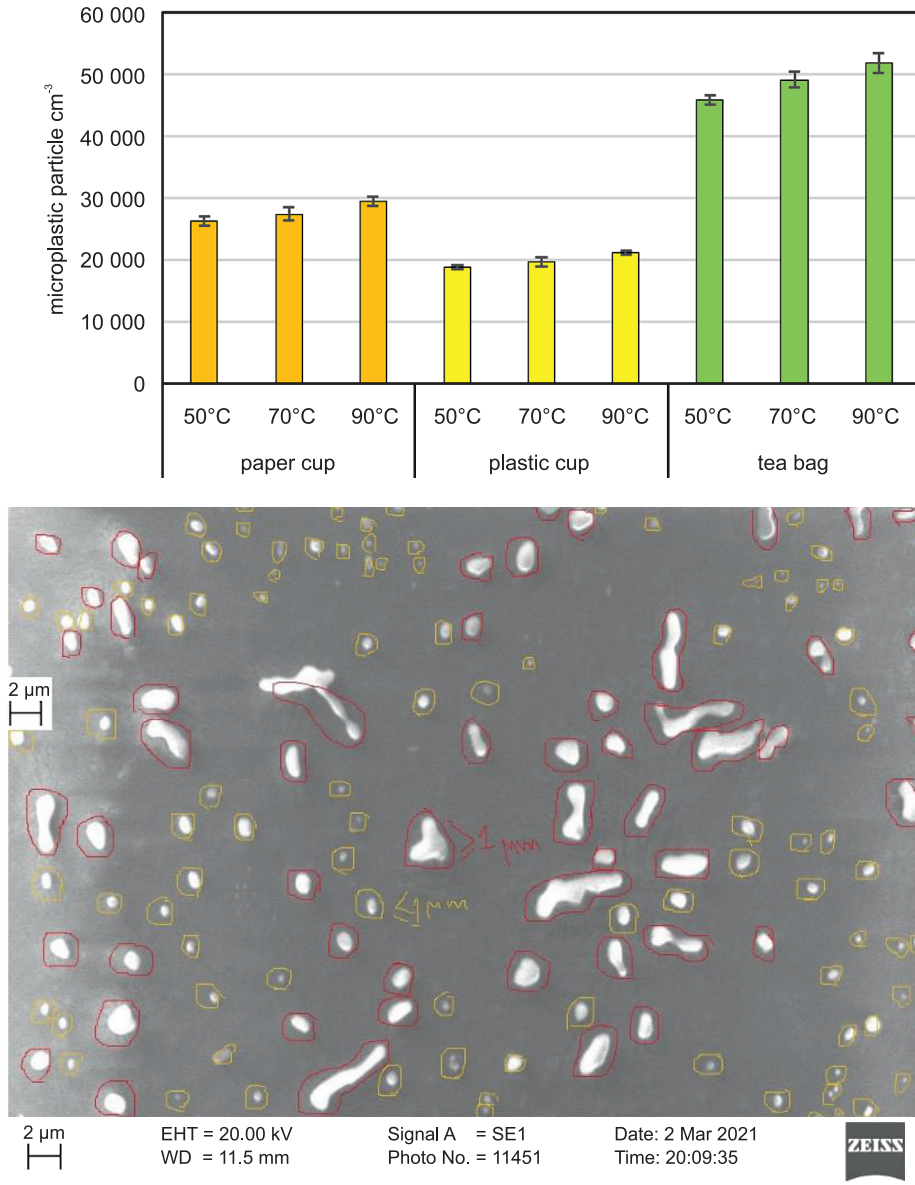

Fig. 2. MP particle count results (upper image, 0.5 µm≤ particle size ≤5 µm, n=3, %RSD=1.3 -3.9) and SEM (lower image, solution volume 200 cm^3, solution temp. 90°C, type: paper cup, those marked in yellow represent microplastic particles smaller than 1 µm, while those marked in red represent microplastic particles 1 µm and larger), image magnification: 10 000X

of 26 281 (±799) particles cm^{-3} was determined at 50°C, while this number was an average of 29 413 (±969) particles cm^{-3} at 90°C. Counts made for plastic cups showed an average of 18 816 (±240) particles cm^{-3} at 50°C,

while this number was an average of 21 077 (±298) particles cm^{-3} at 90°C. The number of microplastic particles that passed from paper and plastic cups into the solution, increased with the effect of temperature and a similar effect was observed for teabags. In the microplastic particle counts processed for teabags, an average of 45 891 (±671) particles cm^{-3} was determined at 50°C, while an average of 51 786 (±1602) particles cm^{-3} was determined at 90°C.

ATR-FTIR analysis results

The broad absorption band at 3320 cm^{-1} in the ATR-FTIR spectrum of MP particles obtained from aqueous solutions containing teabags is stretching vibrations originating from the -OH group and indicates that the particles are in the natural cellulose structure (Figure 3, upper black-lined spectrum).

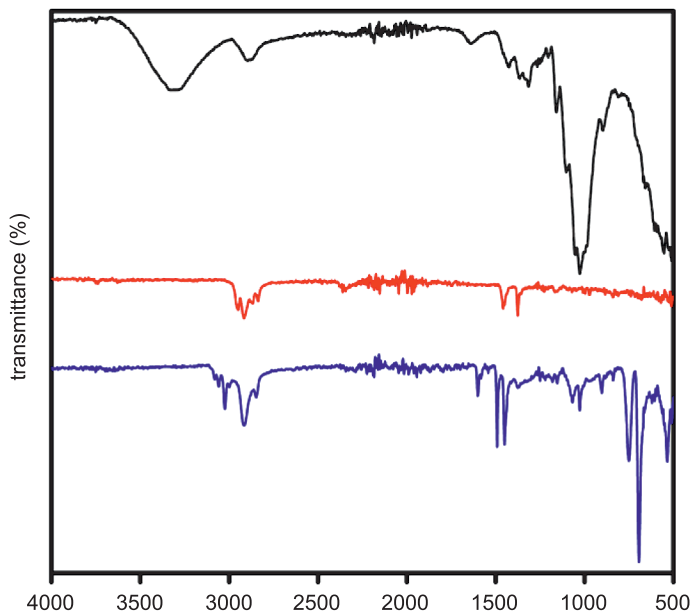

Fig. 3. ATR-FTIR spectrum of particles released from teabags, plastic cups, and paper cups into the aqueous solution

The absorption band corresponding to 1640 cm^{-1} may be caused by bending vibrations of the -OH group or by water molecules absorbed by the cellulose structure. Similarly, these absorption bands are observed in semi-synthetic cellulose structures (Carrillo et al. 2004).

The peak with a frequency of 2880 cm^{-1} indicates -CH stretching vibrations in cellulose structures (Cincinelli et al. 2021).

Cellulose is a natural polymer and consists of linear 1-4 glucose linkages and D-glucopyranose rings attached. The number of units (D-glucopyranose

rings) in this natural polymer structure can be up to 15 000 units. FTIR absorption bands and peaks in natural cellulose structures are also observed in Rayon semi-synthetic cellulose materials. Rayon structure is also called regenerated cellulose (Parajuli et al. 2021).

While natural cellulose fibers are defined by intense peaks at 1430 cm^{-1} frequency caused by symmetrical bending in $-CH_2$ groups, these peak frequencies shift towards 1420 cm^{-1} in semi-synthetic cellulose structures. The rise observed at 1425 cm^{-1} in the spectrum indicates that the MPs released from the teabag structure may be in the semi-synthetic cellulose structure (Figure 3, upper black lined spectrum).

The peak observed at 1105 cm^{-1} in the ATR-FTIR spectrum is essential to distinguish between natural and semi-synthetic cellulose structures. The absorption at this frequency is due to planar asymmetric stretching vibrations in the C-C bonds and is only seen in natural cellulose structures. In our study, the absence of a peak at 1105 cm^{-1} for MP particles released from the teabag indicates that the polymer structure may not be natural cellulose. In the same spectrum, the absorption bands at frequencies 1037 cm^{-1} and 1027 cm^{-1} originate from stretching vibrations in -C-O- bonds, confirming that the polymer structure is semi-synthetic (Cai et al. 2019).

The absorption peaks observed at 807 cm^{-1} and 898 cm^{-1} frequencies are due to symmetrical stretching in the C-C bonds and planar bending vibrations in the $-CH_2$ group, respectively. These peaks are observed in polymers with PP (polypropylene) structure (Chércoles Asensio et al. 2009).

Following the analytical data, it was determined that MP particles released from teabags into aqueous solutions are semi-synthetic cellulose structures containing PP. Some results in studies on this subject in the literature show that teabags have PE (polyethylene), PET (polyethylene terephthalate), PP, or co-polymer structures (Yurtsever 2021).

Transparent plastic cups generally contain polypropylene structure and are marked with the PP5 code. Our study confirmed the polymer structures of MPs released from these plastic cups into aqueous solutions (Figure 3, medium red-lined spectrum).

The absorption bands at frequencies of 2949 cm^{-1} and 2867 cm^{-1} in the spectrum belong to the v-asymmetric and v-symmetric stretching vibrations originating from the $-CH_3$ groups; this confirms the PP structure of MP particles. In the same direction, v-asymmetric and v-symmetric vibrations originating from $-CH_2$ groups in PP structures are observed at 2916 cm^{-1} and 2836 cm^{-1}.

The absorptions in the spectrum at 1458 cm^{-1} and 1356 cm^{-1} (peak shoulder) are δ-in-plane asymmetric and symmetrical bending vibrations of the bonds between carbon and hydrogen atoms in the $-CH_2$ groups. The absorption peaks with the same feature (δ-in-plane asymmetric and symmetrical bending vibrations) originate from the carbon-hydrogen bonds in the $-CH_3$ group in the polymer structure at 1376 cm^{-1}.

The ATR-FTIR spectrum of MP particles released from paper cups into aqueous solutions was recorded (Figure 3, lower blue-lined spectrum). The spectrum's absorption peaks observed at 3083 cm^{-1}, 3060 cm^{-1}, 3022 cm^{-1}, and 2999 cm^{-1} are compatible with the stretching vibrations of the =C-H groups in the benzene ring.

While the peaks at 2917 cm^{-1} and 2848 cm^{-1} in the spectrum consist of symmetric and asymmetric stretching vibrations in the $-CH_2$ structure, the frequencies of 1602 cm^{-1}, 1492 cm^{-1}, and 1451 cm^{-1} belong to the stretching vibrations originating from the C=C bonds in the aromatic structure. The peaks at 1068 cm^{-1} and 1031 cm^{-1} in the spectrum indicate the presence of δ-in-plane bending vibrations in =C-H groups of aromatic structure.

The absorption observed at 754 cm^{-1} with moderate intensity belongs to the δ-plane bending vibrations in the =C-H groups. Specifically, the peaks seen at 693 cm^{-1} and 537 cm^{-1} arise from the aromatic ring's δ- out-of-plane bending vibrations.

LC-MS/MS analysis results of EDC compounds

The aqueous solution samples obtained from plastic cups, paper cups, and teabags were analyzed by LC-MS/MS for 11 phthalate compounds selected to indicate the presence of EDC compounds. As a result of these analyses, besides DMP (dimethyl phthalate), DEP (diethyl phthalate), DBP (dibutyl phthalate), DEHP (bis(2-ethylhexyl) phthalate), BPA (bisphenol A) were also determined quantitatively.

In the LC-MS/MS analyses, the highest concentration values were measured for the DEHP as 1.72 μg dm^{-3}, 1.05 μg dm^{-3} and 1.07 μg dm^{-3} for paper cup, plastic cup, and teabag samples, respectively (Figure 4).

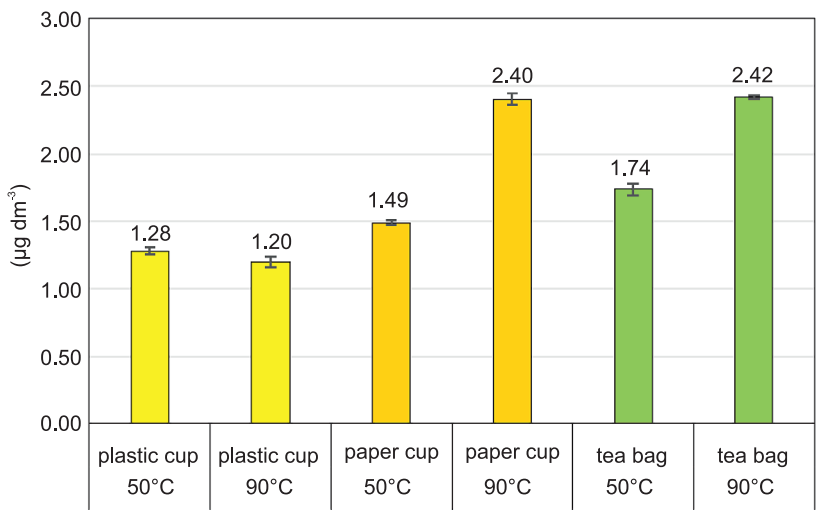

Fig. 4. Total concentrations (μg dm^{-3}) of phthalate compounds (DMP + DEP + DBP + DEHP) released from cups and teabags into water solutions

The total concentrations of phthalate compounds released into aqueous solutions at 90°C were measured as 2.42(±0.02) μg dm^{-3}, 2.40(±0.04) μg dm^{-3} and 1.28(±0.05) μg dm^{-3} for teabags, paper cups, and plastic cups, respectively; in comparison, the concentrations of phthalates decreased except in plastic cups at a lower temperature (50°C) – Figure 4. The total amounts of phthalates measured in water solutions at 50°C were recorded as 1.74(±0.04) μg dm^{-3} in teabag samples while the concentrations detected for paper cup and plastic cup samples were 1.49(±0.02) μg dm^{-3} and 1.28(±0.02) μg dm^{-3}, respectively.

Other critical data obtained from the study shpwed that the phthalate releases change significantly due to increasing temperature. By increasing the water temp. from 50°C to 90°C, the release of phthalate compounds from teabags increased by 42%, while the release rate in paper cups increased by 62%. The temperature increase in the solution in plastic cups did not significantly affect the release of phthalates.

In the analysis of the BPA, the highest concentration determined for plastic cups at 90°C water temperatures was recorded as 0.045 μg dm^{-3}. In contrast, BPA concentrations for paper cups and teabags samples were recorded as 0.006 μg dm^{-3} and 0.011 μg dm^{-3}, respectively.

DISCUSSION

When the results of the analyses were evaluated, the MP particle release from paper cups was higher than from plastic cups, indicating that the idea that paper cups are more harmless needs to be reviewed. In a similar study, Ranjan et al. determined MP particle emissions as 102.3 (±2.1) $x10^6$ particles cm^{-3} (Ranjan et al. 2021). The relatively significant difference between the counts obtained in our study and the MP particle counts in the cited study is because SEM analyses were performed in different particle size ranges (150 nm - 4.277 μm, Ranjan et al.; 500 nm - 5 μm in our study). As a general statement, the amount of MP particles released from all cups and teabags increases with increasing water temperature.

The data obtained from ATR-FTIR analyses were evaluated, and it was determined that MP particles released from teabags into aqueous solutions are semi-synthetic cellulose structures containing PP. Some results in studies on this subject in the literature show that teabags have PE (polyethylene), PET (polyethylene terephthalate), PP, or co-polymer structures (Yurtsever 2021).

Interpreting the ATR-FTIR analytical results, it was confirmed that the structures of the MPs that passed from the plastic cups to the aqueous solution under the specified experimental conditions were PP (Mecozzi et al. 2016).

We analyzed the data we collected from the spectrum. It was determined that the MP particles released from paper cups to the aqueous solutions corresponed to the structure of PS (polystyrene) polymer (Chércoles Asensio et al. 2009, Castelvetro et al. 2021, Fernández-González et al. 2021).

LC-MS/MS analyses of analytes, phthalates (11 compounds), and BPA were conducted in our study. Out of 12 selected analytes, we quantified DMP, DEP, DBP, DEHP, and BPA in our research. In the previous studies performed and reported in the literature, the detection frequencies of compounds were compared; the presence of DBP at 67.6%, DEHP at 61.7%, DEP at 47.1%, and DMP at 36.9% of the samples were quantitatively determined (Luo et al. 2018)

These data are consistent with the results of our study (Otero et al. 2015, Salazar-Beltrán et al. 2018). The upper limit of DEHP in drinking water stored in plastic containers has been determined by the World Health Organization as 8.0 μg dm^{-3}.

Although there is no regulation regarding the limit values in hot drinks, the highest value determined for DEHP in our study (1.72 μg dm^{-3}, 90°C, paper cups) is below the upper limit value determined for drinking water in plastic bottles (Organization 24 April 2017).

It was observed that BPA emission in plastic cups increased significantly with increasing water temperature (from 50°C to 90°C); however, this temperature change did not considerably fluctuate the BPA release in paper cups and teabag samples. The upper limit allowed for BPA in beverages has been set by the European Commission as 50 μg dm^{-3} (Khan et al. 2021).

The amount of MP and chemical additives such as phthalate and bisphenol A released from plastic cups, paper cups and teabags into hot water is critical for health. The release of MP increases in direct proportion to water temperature. As a result of using disposable cups and teabags decomposing over time, MP mixes into soil, water, and air. Therefore, exposure to MP and chemical additives is expected to be much higher than estimated.

Considering the effects of MPs on the environment and human health, the European Union banned the use of products such as plates, forks, knives, glasses, earbuds and straws made of disposable plastic on July 3rd, 2021 (Congar 2021). And in Turkey, on 1 January 2019, paid plastic bags in stores were introduced to reduce plastic waste (İbrahim, Salih 2020). Even though these initiatives show that the necessary sensitivity to MPs' effects on the environment and human health has begun to emerge, it is apparent that there is a need for new, more comprehensive regulations regarding both the use and production of single-use plastics.

CONCLUSIONS

The number of MPs released from plastic cups, paper cups, and teabags increase significantly at elevated temperature of liquids. In our study, water solutions were prepared to mimic the actual consumption conditions of beverages.

In general, detected EDC concentrations in these water solutions were affected by elevated temperatures of solutions and increased positively by high solution temperatures.

Teabags could be made from polymeric substances, contrary to natural cellulose, or may contain semi-synthetic polymeric structures. This issue should be considered seriously as MP particle emissions from teabags into hot water solutions exceed the release of MPs from plastic cups and paper cups.

To evaluate the overall effect of the MP's intake from plastic cups, paper cups, and teabags used daily in the consumption of hot beverages on human health, further studies should be designed and conducted.

Author contributions

N.T. – designed research. K.S., S.C. – conducted experiments. N.T., K.S., S.C. – analyzed data, and wrote the manuscript. All authors read and approved the manuscript.

Conflicts of interest

The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data, in the writing of the manuscript, or in the decision to publish the results.

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