# Microplastics and phthalate esters release from teabags into tea drink: Occurrence, human exposure, and health risks

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## Abstract

Tea, the most common infusion worldwide, is usually sold in teabags due to the ease of usage. The increase in the use of plastic materials in the food packaging industry has led to an increase in released contaminants, such as microplastics (MPs) and phthalates (PAEs), in various food products including teabags. In this research, the abundance and features of MPs as well as PAEs concentration were investigated in 45 teabag samples of different Persian and German brands. The abundance of MPs in the Persian and German teabag samples were averagely 412.32 and 147.28 items/single teabag, respectively. Also, average PAEs levels in the Persian and German teabag samples were 2.87 and 2.37 mg/g, respectively. The predominant size category of MPs was relalated to 100–250 μm. Fibers and transparent were the dominant shape and color of detected MPs in teabags, respectively. Polyethylene (PE) and nylon were the most common MP polymer types. The most prominent PAEs congeners in teabag samples were diethylhexyl phthalate (DEHP) and diisobutyl phthalate (DiBP). Furthermore, MP exposure hazard through tea ingestion was analyzed for three age groups: Teens, adults, and children. DEHP showed the cancer risk (CR) for age groups of children, teens, and adults. The findings of this research indicated that high MPs and PAEs levels are released from teabags into tea drinks. Considering a daily drinking of a volume of 200–400 mL tea, 648–1296 MPs may enter the human body. Thus, tea prepared with teabag-packed herbs may pose a significant health risk for consumers.


## 1. Introduction

The production and usage of plastics have continuously increased through the years due to their useful properties, such as high stability, lightness, and flexibility (Okoffo et al., 2021), which make them suitable for multiple uses. More specifically, plastic consumption has enhanced from 1.5 million tons in 1950 to 348 million in 2017 (Ito et al., 2019; Petersen and Hubbart, 2021). Plastic debris less than 5 mm are called microplastics (MPs), which are divided into primary and secondary groups according to their origin. Primary MPs are manufactured small microscopic particles, while secondary particles are produced from larger plastic particles through physical and chemical decomposition (Akhbarizadeh et al., 2021b; Dobaradaran et al., 2018). Phthalate esters (PAEs) as the largest plasticizers group are applied in plastic products, such as those based on polyethylene terephthalate (PET), polyvinyl chloride (PVC), and polyethylene (PE), with the objective of improving their elasticity and performance (Arfaeinia et al., 2019; Mohammadi et al., 2022; Takdastan et al., 2021). Nowadays, a great number of foods and consumable products are stored in plastic containers due to their ease of transportation and prolonged shelf life (Jadhav et al., 2021). Thus, the widespread application of plastics in the food packaging industry could potentially lead to the contamination of consumable products (Fadare et al., 2020; Xu et al., 2019). Food processing, plastic containers, and packaging may also results in the contamination of beverages and food with chemical additives, such as PAEs (Dobaradaran et al., 2020).

The MPs presence in numerous environmental samples, including bottled water (Schymanski et al., 2018), tap water (Kosuth et al., 2018), cold and energy drinks (Shruti et al., 2020), table salt (Nithin et al., 2021), beer (Liebezeit and Liebezeit, 2014), sugar (Afrin et al., 2022b), canned fish (Akhbarizadeh et al., 2020a), seafood (fish and shellfish) (Panebianco et al., 2019), indoor dust (Kashfi et al., 2022), runoff (Hajiouni et al., 2022), wastewater effluents (Hajji et al., 2022; Takdastan et al., 2021), landfill leachate (Mohammadi et al., 2022), air (Akhbarizadeh et al., 2021b), and personal protective equipment (Akhbarizadeh et al., 2021a; De-la-Torre et al., 2022) has been reported. Also, the occurrence of PAEs in bottled water (Abtahi et al., 2019; Wang et al., 2021), tap water (Abtahi et al., 2019), beverages (Arfaeinia et al., 2020; Caldeirão et al., 2021; Kiani et al., 2018), dough (Kouhpayeh et al., 2017), fish (Cheng et al., 2013; Lee et al., 2019), urban runoff (Hajiouni et al., 2022), landfill leachates (Mohammadi et al., 2022), sediment (Arfaeinia et al., 2019), wastewater effluents (Takdastan et al., 2021), and personal protective equipment (De-la-Torre et al., 2022) have been investigated in the recent studies. To date, limited researches have considered the MPs level (Afrin et al., 2022a; Busse et al., 2020; Hernandez et al., 2019) and PAEs concentration (Alnaimat et al., 2020; Yurtsever, 2021) in the tea drinks prepared using teabags.

In many countries, drinking tea is a tradition, while teabags are mostly prefered over bulk tea leaves (Afrin et al., 2022a). Teabags are often made of plastic or have a plastic cover. Thus, commercial tea products can be a source of MPs and PAEs due to the packaging. Placing teabags at brewing temperature (95°C or higher) increases the release of MPs and PAEs into tea infusions (Sewwandi et al., 2022). It was reported that cell uptake and displacement of different types of MP particles across the mammalian intestine may lead to tissue damage, fibrosis, and inflammatory and cytotoxic impacts on lung epithelial of human (Prata et al., 2020; Schirinzi et al., 2017). Also, PAEs have been illustrated to cause harmful effects on human health under chronic and high exposure, such as infertility, sexual abnormalities, asthma, nephron-and hepato-toxicity, high blood pressure, type II diabetes, obesity, and neurological disorders, as well as cancer (Benjamin et al., 2017; Giuliani et al., 2020).

Considering the issue that teabags may release MPs and PAEs into tea, it is necessary to investigate the possible health hazards of noted contaminants for humans. As no former research has simultaneously studied MPs and PAEs release from teabags into tea infusion, in this work, the MPs and PAEs presence in different commercially available Persian and German teabags was investigated. Also, the estimated daily intake (EDI) of MPs for teabag consumers was reported for the first time in this study. The specific aims of this study were to 1) determine the abundance and features (morphology, size, color, and type of polymer) of MPs released from different Persian and German teabags into tea; 2) determine the $\sum_7$PAEs levels released from different Persian and German teabags into tea; and 3) determine the EDI of MPs and human health risk of PAEs.

## 2. Materials and methods

## 2.1. Sample collection

Three samples of 15 different commercial brands of teabags, including 10 Persian brands and 5 German brands (total of 45 samples), were collected from different stores in December 2021. For ethical reasons, the brands' names of teabags are not mentioned here. The Persian brands are presented as $A_1$-$A_{10}$ and the German brands as $B_1$-$B_5$. The information on each teabags brand, including the place of production and packaging, is shown in Table S1. All provided teabag samples were individual packaging that are usually used for one cup.

## 2.2. Sample processing

The extraction of MPs was done based on the previous study with a few modifications (Hernandez et al., 2019). To determine MP release from each empty teabag, the plastic bag was cut with metal scissors and the tea leaves were separated from them. The empty teabags were thoroughly rinsed 3 times with deionized water to eliminate plastic and tea particles, then dried with a stream of nitrogen. Each empty teabag was located in a 400 mL glass beaker and 250 mL of deionized water was added at a temperature of 95°C for 5 min. Then the samples were covered with aluminium foil and held at room temperature for 3 h. The prepared samples were filtered by vacuum filtration via a polytetrafluoroethylene polymer (PTFE) memberane filter (Whatman, Maidstone, Kent, UK, pore size of 0.45 μm). Finally, the filters containing MP pieces were dried at room temperature in the glass Petri dishes covered with aluminium foil.

The extraction of PAEs in teabags was performed according to the method of the former study with small modifications (Troisi et al., 2019). Hundred mL of each sample prepared by the empty teabags for the extraction of MPs was filtered by PTFE filters with a pore size of 0.45 μm. Twenty mL of dichloromethane (Merck, Germany) was added to samples for PAEs extraction by separating funnel (liquid-liquid extraction). It should be noted that 20 μL benzyl benzoate (Merck, Germany) with concentration of 60 μg/L was added to the samples at the beginning of the work-up as internal standard. Then each sample was stirred for 5 min. Finally, the extracts were concentrated to 2 mL under a gentle flow of nitrogen and analyzed by a gas chromatography-mass spectrometry (GC-MS).

## 2.3. MPs analysis

By a binocular microscope (A. KRÜSS optronic, Germany) with up to 400X magnification, the physical features of MPs, such as size, shape, and color, were identified. The MPs characteristics including non-shininess, elasticity, identical thickness, homogeneous color, and inorganic surface structure were tested using a probe. A hot probe test was applied to ensure the plasticity of the MP particle in some cases (Hidalgo-Ruz et al., 2012). The particle size was evaluated through the probe diameter and recorded. MPs were categorized into 3 shapes including fibers, films, and fragments (Hartmann et al., 2019).

To detect the MPs chemical composition, micro-Raman (Lab Ram HR, Horiba, Japan) was applied. A laser irradiated at 785 nm was used as the excitation source and the spectral of Raman was gained at 400–1800 cm$^{-1}$. Eventually, by comparing the obtained spectra with the spectral libraries of plastic particles for Raman microscope, the particles polymer composition was determined.

## 2.4. PAEs analysis

GC-MS device (Agilent Technologies, Avondale, PA, US) equipped with a quadrupole mass spectrometer was used for PAEs analysis. The details of PAEs analysis by GC-MS are mentioned in the supplementary file.

## 2.5. Human exposure to MPs and PAEs

The EDI of MPs and PAEs through consumption of teabag was quantified for evaluating the exposure of 3 age groups including children, teenagers, and adults to these contaminants. EDI was estimated based on the following equation (Enyoh et al., 2021; Rakib et al., 2021):

$$EDI_{Ingestion} = \frac{C \times DI \times EF \times ED}{BW \times AT}$$

1

Where C is the number of MPs (items/single teabag) and PAEs concentration (mg/L) released from the teabag. DI is the average daily intake of tea (L/day). It should be noted that each teabag is usually used for one cup, which is considered a 200 mL serving. The average consumption of tea per person was considered two cups per day (400 mL) (EFSA, 2013). ED is the exposure duration (year), EF is exposure frequency (day/year), AT is the averaging time (days), and BW is body weight (kg); and the unit of EDI is mg/kg/day. The values of EF, ED, BW, and AT for three age groups including children (with the age range of 0–10 years), teenagers (with the age range of 11–20 years), and adults (with the age range of 21–72 years) were considered based on a former study in Iran (Miri et al., 2018).

The hazard quotient (HQ) and cancer risk (CR) were used to estimate the non-carcinogenic and carcinogenic risks of PAEs compoundsby, respectively. The hazard quotient (HQ) indicates the non-cancer risk of PAEs and was determined based on the following equation (USEPA, 2002):

$$HQ = \frac{EDI}{RfD}$$

2

Where RfD (mg/kg-day) is the oral reference dose of PAEs compounds; that was gained from the database of the Integrated Risk Information System, USEPA (USEPA, 2002). RfD values for dibutyl phthalate (DBP), diethyl phthalate (DEP), butyl benzyl phthalate (BBP), dimethyl phthalate (DMP), dioctyl phthalate (DOP), and diethylhexyl phthalate (DEHP) are 0.1, 0.8, 0.2, 0.1, 0.01, and 0.02 mg/kg-day, respectively. The value of HQ less than 1 shows an acceptable level without an increment of adverse health effects, while the value of HQ greater than 1 indicates a probability of non-carcinogenic health effects for humans (USEPA, 2002).

The CR values were estimated as follows (USEPA, 2001):

$$\mathrm{CR} = \mathrm{EDI} \times \mathrm{SF}$$

3

Where SF is the slope factor of PAEs compounds (mg/kg/day). SF values for BBP and DEHP are 0.0019 and 0.014 $(mg/kg\text{-}day)^{-1}$, respectively. The acceptable value of carcinogenic risk is between $10^{-6}$ and $10^{-4}$ (USEPA, 2001).

## 2.6. Quality control (QC)/quality assurance (QA)

Before the experiment and MP analysis, ethanol and paper wipes were used to clean all tools and work tables. To avoid the contamination of the sample, glass containers were applied during all experiments. Furthermore, before usage, the glass vessels were attentively washed with ultrapure water. In all stages of preparation and extraction, the samples surface was concealed with the aluminum foil. Cotton coats and nitrile gloves were used during all analytical procedures steps. In addition, during the sample analysis, five control Petri dishes with distilled water were applied to check contamination of airborne in the lab. To control the background contamination, blank samples were provide for each set of samples. In the blank samples, a few numbers of MPs were found and the values were subtracted from the number of counted MPs to perform corrections to all samples. The PAEs calibration standards were applied for the calibration of the instrument. The obtained calibration curves for the PAEs congeners illustrated good linearity ($R^2$: 0.99–0.999). The PAEs compounds illustrated good recovery in the range of 87.8-104.3% with good precision (%RSD: 1.9–9.4%). A signal to noise ratio of 3 were used to identify the limit of detection (LOD). The PAEs compound abbreviations, LOD, the limit of quantification (LOQ), and recovery percent (R%) for the quantified PAEs are presented in Table S2. The levels of DEP, DMP, diisobutyl phthalate (DiBP), BBP, DBP, DOP, and DEHP that were detected in the procedural blanks were deducted from the results.

## 2.6. Statistical analysis

IBM SPSS 20 and Microsoft Excel 2016 software were utilized to analyze MPs and PAEs in the teabags with different brands. Descriptive statistics were used to describe and analyze the levels of MPs and PAEs. After determining the normality of data, the difference among the levels of MPs and PAEs in the Persian and German teabags was determined by the T-test analysis. Statistical significance was accepted at $p < 0.05$.

### 3. Results and discussions

## 3.1. MPs in teabags

## 3.1.1. MPs abundance

MP particles were observed in all teabag samples. A total of 100 fiber particles were observed in the five control samples, which were deducted from the counted MP particles. Averagely the MPs numberin different Persian ($A_1$-$A_{10}$) and German ($B_1$-$B_5$) tea bag brands were in the range of 73.6-1446.8 and 55.6-323.2 items/single teabag, respectively, as shown in Fig. 1 and Table S3. Based on the Fig. 1, the lowest MPs abundance in the teabag samples was related to the $B_3$ brand with a level of 55.6 items/single teabag, and the highest MPs abundance was found in the $A_3$ brand with a value of 1446.8 items/single teabag. These results showed that the use of plastic bags in the packaging of teas causes the leachate of MPs into the tea. A significant difference was detected between the abundance of MPs in the Persian and German teabag brands (p-value < 0.05). The difference among the MPs abundance in the Persian and German brands can be due to the characteristics of product packaging and the amount of plastic used in the packaging of teabags. The MPs abundance and their characteristics in this research are compared with other researches as presented in Table 1. In a study in Bangladesh, the average levels of MPs released from intact teabags (with tea leaves) and empty teabags (without tea leaves) were reported as ≈ 50 and ≈ 65 items/kg, respectively (Afrin et al., 2022a). In another study in Canada, the MPs level released from a plastic teabag into tea infusion was reported ≈ 11.6 billion/one cup (Hernandez et al., 2019). In a study in Mexico, the MPs abundance in the cold tea samples was reported as 11 items/mL. According to the findings of the mentioned study, the particles number in drinks depends on the packaging material type of food products (Shruti et al., 2020). The difference in the MPs number released from teabags into tea infusions can be due to the difference in the plastic materials used for teabag packaging and other factors that affect the degradation of plastic packaging, such as aging, temperature, and external mechanical forces, as well as analytical parameters such as the samples number and instrumental sensitivity applied for MPs identification (Chen et al., 2023; Jadhav et al., 2021). The teabags are usually kept in a paper package that is probably separated by a layer of plastic. Also, MPs may stick to teabags in the packaging process of teabags (Tsochatzis et al., 2020). Increasing the damage of teabags, the temperature of the water, and boiling time may enhance the release of MP particles (Kim et al., 2022). The migration of MPs from teabags into tea increases at higher temperatures, especially at temperatures higher than 95°C, due to the thermal degradation of polymers (Hernandez et al., 2019). Thus, the MPs release from plastic containers when they are exposed to hot beverages and food should be more considered.

Table 1
Levels and characteristics of MPs in teabags, tea leaves, and tea infusions reported worldwide.

| Study area (n = Sample number) | Sample type | MPs abundance | Unit | Dominant size range (μm) | Major shapes | Major color | Major polymer types | Ref |
|---|---|---|---|---|---|---|---|---|
| Bangladesh (n = 5) | Intact teabag | ≈ 50 | items/kg | 23–60 | Fiber | Blue | ABS[1] | (Afrin et al., 2022a) |
| | Empty teabag | ≈ 65 | items/kg | 23–60 | Fiber | Brown | EVA[2] | |
| Canada (n = 4) | Teabag | ≈ 11.6 billion | items/one cup | 1-150 | Fiber | Transparent | Nylon | (Hernandez et al., 2019) |
| Ireland (n = 6) | Teabag | - | - | 951–2496 | - | - | PP[3] | (Xu et al., 2021) |
| South Korea (n = 6) | Teabag | - | - | - | Fiber | - | Nylon | (Kim et al., 2022) |
| Mexico city (n = 4) | Cold tea | 11 | items/L | 100–500 | Fiber | Blue | Nylon | Shruti et al., 2020) |
| China (n = 3) | Teabag | - | - | 620–840 | Fragment | Transparent | PET[4], PP | (Mei et al., 2022) |
| China (n = 4) | Tea leave | 200–500 | items/g | - | Fiber | Transparent | PE[5], PET | (Li et al., 2022) |
| **Iran (n = 30)** | **Teabag** | **412.32** | **items/single teabag** | **100–250** | **Fiber** | **Transparent** | **Nylon, PE[3]** | **This study** |
| **Germany (n = 15)** | **Teabag** | **147.28** | **items/single teabag** | **100–250** | **Fiber** | **Transparent** | **Nylon, PE** | |

1: Acrylonitrile butadiene styrene; 2: Ethylene-vinyl acetate; 3: Polypropylene; 4: Polyethylene terephthalate; 5: polyethylene

## 3.1.2. Physical characteristics of MPs

The detected MPs in the teabag samples was classified into five various size categories as illustrated in Fig. 2 (a) and Table S4. The size distribution of identified MPs in the teabags varied from 20 to 5000 μm. Among detected MPs, 41.09% were between 100–250 μm, 26.65% between 250–500 μm, 20.61% between 500–1000 μm, 5.7% between 1000–5000 μm, and 5.16% between 20–100 μm. The most abundant detected MPs in all teabag brands were associated with the size class of 100–250 μm (averaging 41.09% in all samples), except for $A_{10}$ and $B_3$. Based on the Table 1, the most abundant size category of identified MPs in teabag samples in China (Mei et al., 2022) and Bangladesh (Afrin et al., 2022a) were reported as 620–840 μm and 23–60 μm, respectively. The larger size of MPs may fragmente to smaller size particles due to transportation process and water temperature (Zhou et al., 2023). The decrease of the size of MPs enhances their surface area and consequently increases their reactivity and degradation (Jadhav et al., 2021; Klein et al., 2018). The tiny particles risk is higher compared to bulk plastics. The higher surface area and smaller size allow for easy cell membrane penetration due to physical damage or adsorption onto the cells surface (Agathokleous et al., 2021; Mateos-Cárdenas et al., 2021; Shi et al., 2022).

Figure S1 (a) and Table S4 illustrate the MPs shapes percentage and their number found in the different Persian and German teabag brands. Images of identified MPs are also shown in Fig. S2. Fiber was the most abundant shape of MP (99.99%) among the investigated particles and only 9 detected MPs were in the shape of film (0.01%). The result of this study was similar to former studies in Bangladesh (Afrin et al., 2022a), Canada (Hernandez et al., 2019), and South Korea (Kim et al., 2022) as shown in Table 1. Another study in Mexico has also shown that fibers are the most abundant form of MPs in cold tea (Shruti et al., 2020). Microscopic images of empty teabags revealed that the bags are composed of an irregular and dense network of cellulose fibers (Xu et al., 2021). The different factors and processes of degradation can cause the abrasion of plastic materials to form plastic particles (Jadhav et al., 2021).

The color percentages and number of the detected MPs are shown in Fig. S1 (b) and Table S4. The identified MPs color in the teabags followed the order of white/transparent (99.1%) > black (0.63%) > red/pink (0.24%) > blue/purple (0.03%). The dominant color of detected MPs was white/transparent that is in accordance with the previous studies in Canada (Hernandez et al., 2019), and China (Mei et al., 2022). However, in a study in Bangladesh, brown and blue were reported as the most abundant color of MPs released from teabags (Afrin et al., 2022a) (Table 1). The color of MPs helps to identify the source of their production. The preseance of MPs with white/transparent color in the teabags can be because the light polymers usage for their packaging. The release of MPs from plastic packaging into beverages or food mostly associate with their source influencing, MPs characteristics such as color and polymer types (Jadhav et al., 2021; Kedzierski et al., 2020).

## 3.1.3. Polymer types of MPs

Twentynine MP particles were randomly selected and analyzed using Raman Spectroscopy to identify the type of polymer. Figure 3 illustrates the abundance of different polymer types detected through Raman spectra. The raman spectra of some detected MPs polymers released from teabags are also shown in Fig. S3. The identified polymer types of MPs were PE (53.9%), nylon (42.3%), and polycarbonate (PC) (3.8%). In a previous study in Bangladesh, the dominant polymer types of MPs released from intact and empty teabags into tea infusions were ethylene-vinyl acetate (EVA) and acrylonitrile butadiene styrene (ABS), respectively (Afrin et al., 2022a). While, PET and PP were the dominant types of polymer released from teabags in China (Mei et al., 2022). In a similar study in Canada, approximately billions of nylon and PET particles were found in a single cup of tea. According to their results, a nylon teabag starts to degrade in hot water (at 95°C), thus leading to the release of particles into the tea infusion(Hernandez et al., 2019). In a study in Mexico, the polymer types of MPs released from teabags into tea infusion were nylon and PET (Shruti et al., 2020). In another study in Turkey, polyester, PE, PP, and PP-PE blends were the detected MPs polymer types in teabags (Yurtsever, 2021). The MPs polymer composition indicates the origin of their production (Lehmann et al., 2021). The major polymer type of identified MPs in foods packaged with plastic materials were PET, PP, and nylon (Jadhav et al., 2021). Nylon and PE are utilized in cellulose and synthetic textiles used for the packaging of teabags (Shruti et al., 2020). The beverage and food packaging are coated with PE polymer and a layer of cellulosic material (Schymanski et al., 2018). PC is also extensively applied in the producing industries, especially packing (Antonakou and Achilias, 2013).

## 3.2. PAEs levels in teabag

The PAEs leachates from teabag samples are presented in Fig. 4 (a) and Table S5. The highest and lowest average PAEs concentrations were detected in $A_1$ and $B_3$ brands with levels of 5.49and 1.32 mg/g (9.11 and 2.19 mg/L), respectively. In this study, 7 PAEs congeners including DEP, DMP, DBP, DiBP, DEHP, BBP, and DOP were identified. As shown in Fig. 4 (b) and Table S5, DEHP and DiBP were the dominant PAEs dectected. No statistically significant differences were detected among the average levels of PAEs in different brands of the Persian and German teabag samples ($p\text{-value} > 0.05$, Table S5). The findings of this study indicated that the PAEs release rate is significant. The levels and congeners of detected PAEs in this study are compared with other studies, as shown in Table 2. DMP, DBP, and BBP were the detected PAEs congeners leachate from teabag into tea infusion in a study in Spain (Alnaimat et al., 2020). In a study in India, BBP, DBP, and DEHP were detected in teabags (Yadav et al., 2017). In another study in Canada on the available teabags across the global market, DBP, BBP, and DEHP were found in teabag samples (Carr Kinnear et al., 2021). The PAEs concentration in the leaves and infusions of tea was also investigated in former studies. For example, in a study in China and UK, the total levels of five PAEs including DEP, DMP, DBP, DiBP, and DEHP in the tea leaves and their infusions were in the range of 1.13–3.73 and 0.009–0.78 mg/kg, respectively. DiBP, DBP, and DEP were the dominant PAEs congeners in the different tea leaves and their infusions (Du et al., 2016). PAEs occurrence in teabags can be due to the materials used in making the bags and other compounds, such as artificial essential oils, color, and flavorings in the tea production line (Giuliani et al., 2020). A high temperature can increase the PAEs leaching via teabags. Thus, the brewing temperature and time have a considerable effect on the leaching of PAEs (Carr Kinnear et al., 2021). Overall, the concentration of PAEs in water is low due to their fat-soluble, but the high water temperature for brewing tea largely overcomes its low solubility in water. Also, contact of tea leaves with packaging plastic can be the most important source of PAEs (Santana-Mayor et al., 2021).

Table 2
Concentrations and congeners of PAEs in teabags in this study compared with teabags, tea leaves, and tea infusions in other locations of worldwide.

| Study area (n = Sample number) | Sample type | PAEs congeners (min-max) | ∑PAEs level (min-max) | Unit | Ref |
|---|---|---|---|---|---|
| Spain (n = 9) | Teabag | BBP[1] (< LOD*-0.4) | - | mg/L | (Alnaimat et al., 2020) |
| | | DBP[2] (10-51.7) | | | |
| | | DMP[3] (< LOD-51.9) | | | |
| | Tea infusion | BBP (< LOD-0.62) | - | mg/L | |
| | | DBP (32.7-562.2) | | | |
| | | DMP (< LOD-2.9) | | | |
| China and UK (n = 25) | Tea leave | DMP (< LOD-0.7) | 1.13–3.73 | mg/kg | (Du et al., 2016) |
| | | DEP[4] (< LOD-1.42) | | | |
| | | DiBP[5] (0.1–1.06) | | | |
| | | DBP (0.11–1.73) | | | |
| | | DEHP[6] (0.02–0.53) | | | |
| | Tea infusion | DMP (< LOD-0.1) | 0.009–0.89 | mg/kg | |
| | | DEP (< LOD-0.23) | | | |
| | | DiBP (< LOD-0.39) | | | |
| | | DBP (< LOD-0.37) | | | |
| | | DEHP (< LOD-0.17) | | | |
| China (n = 6) | Tea leave | DEP (3.2–18.3) | 69.7–2244 | mg/kg | (Liu et al., 2016) |
| | | DBP (14.3–50.3) | | | |
| | | DMP (28.3-271.7) | | | |
| | | DiBP (7.7-158.3) | | | |
| | | DEHP (10.7-353.3) | | | |
| | | DMEP[7] (< LOD-1203) | | | |
| | | DNP[8] (< LOD-246.9) | | | |
| | | DPrP[9] (< LOD-130.2) | | | |
| Canada (n = 6) | Teabag | DMP (< LOD-56) | - | ng/g | (Carr Kinnear et al., 2021) |
| | | DEP (< LOD-144) | | | |
| | | DBP (42–171) | | | |
| | | BBP (7-244) | | | |
| | | DEHP (6-116) | | | |
| | | DOP (< LOD-101) | | | |
| | Tea leave | DMP (31–32) | - | ng/g | |
| | | DEP (38–48) | | | |
| | | DBP (17–21) | | | |
| | | BBP (24–25) | | | |
| | | DEHP (27–27) | | | |
| | | DOP[10] (25–27) | | | |
| India (n = 100) | Teabag | DBP (< LOD-0.18) | - | mg/kg | (Yadav et al., 2017) |
| | Tea infusion | DBP (< LOD-0.023) | - | mg/kg | |
| **Iran** | **Teabag** | **DMP (0.08–0.76)** | **1.48–5.49** | **mg/g** | **This study** |

| Study area (n = Sample number) | Sample type | PAEs congeners (min-max) | ∑PAEs level (min-max) | Unit | Ref |
|---|---|---|---|---|---|
| (n = 30) | | DEP (< LOD-0.6) | | | |
| | | DiBP (0.19–1.84) | | | |
| | | DBP (0.01–0.49) | | | |
| | | BBP (< LOD-0.14) | | | |
| | | DEHP (0.37–4.55) | | | |
| | | DOP (0.01–0.22) | | | |
| Germany (n = 15) | Teabag | DMP (0.11–0.57) | 1.32–3.73 | mg/g | |
| | | DEP (0.06–0.2) | | | |
| | | DiBP (0.06–0.49) | | | |
| | | DBP (0.01–0.49) | | | |
| | | BBP (0.01–0.07) | | | |
| | | DEHP (0.53–2.45) | | | |
| | | DOP (0.02–0.18) | | | |

1: Benzyl butyl phthalate; 2: Dibutyl phthalate; 3: Dimethyl phthalate; 4: Diethyl phthalate; 5: Diisobutyl phthalate; 6: Diethylhexyl phthalate; 7: Dimethoxyethyl phthalate; 8: Dinonyl phthalate; 9: Dipropyl phthalate; 10: Dioctyl Phthalate

* Limit of detection

## 3.4. Corrolation among MPs and PAEs

Due to originate of PAEs from plastic particles, the relationship among the abundance of released MPs and levels of PAEs leached from teabag was determined as presented in Fig. 5. As shown, a positive relationship among abundance of MPs and PAEs concentration ($r = 0.47$, $p < 0.05$) was found. Significant relationships among MPs number and PAEs levels have been also found in urban runoff and landfill leachate samples investigated in Iran (Hajiouni et al., 2022; Mohammadi et al., 2022), sediments investigated in Mexico (Ramirez et al., 2019), and seawater investigated in China (Zhang et al., 2019). The polymer pore size, the plastic materials structure, and the additive's physical-chemical features may affect the PAEs migration from plastic materials (Xu et al., 2020). Generally, the findings of this study indicated that the quantitative calculation of PAEs can be associated with MPs contamination.

## 3.5. Human exposure to MPs

Identifying the routes of human exposure to plastic debris in the environment is essential for health risk assessment of MPs (Fadare et al., 2020). Oral ingestion of MPs may enhance the MPs' bioavailability in the digestive tract causing harmful effects on the human body (Sewwandi et al., 2022). According to the annual or daily consumption of various beverages and food, body weight, and MPs content, the human MPs intake can be determined numerically (Cox et al., 2019; Zuccarello et al., 2019). The daily MP intake for 3 age groups including children, teenagers, and adults via 400 mL (2 cups) of tea infusion prepared using teabags are presented in Table 3. The highest exposure risk was found for children in the case of all teabag brands. In the Persian brands, EDI values for children in the case of $A_3$ and $A_5$ brands were the highest with values of 259 and 245 items/kg-BW/day. In the German brands, EDI values for children in the case of $B_5$ and $B_1$ brands were the highest with values of 57.9 and 36.3 items/kg-BW/day. The higher exposure risk for children can be due to the lower body weight than other age groups. The EDI values via 600 mL (3 cups) of tea infusion prepared by the Persian and German teabags were also estimated for children, teens, and adults and were in the range of 14.93-388.54, 5.38-140.12, 1.04–27.27 items/kg-BW/day, respectively. Thus, increasing tea consumption prepared by teabags can significantly increase MPs intake. In a study in Malaysia, the estimated EDI value for MPs ingestion via bottled water for adults was between 0.068 and 0.19 items/kg-BW/day, while for children this value was between 0.089 and 0.25 items/kg-BW/day (Praveena et al., 2022). In a review, the EDI values of MPs via plastic and glass bottled water consumption were determined as ≈ 85 and ≈ 220 items/kg-Bw/day, respectively (Akhbarizadeh et al., 2020b). In another review, the EDI value of MPs via milk consumption was reported as 6.6 items/kg-BW/day. The EDI values of MPs via consumption of other drinks, such as beer and beverages, were lower than 1 item/kg-BW/day (Lin et al., 2022). Also, according to the results of a study in China, people may ingest 37613–89294 MP particles per year due to one plastic cup usage of water every 4–5 days (Zhou et al., 2023). However, there is yet no evidence of MPs exposure and health impacts through tea consumption that should be considered in further studies.

Table 3
Estimated daily intake (EDI) of MPs (items/kg-Bw/day) for teabag consumers.

| Country | Teabag brand | Children | Teens | Adults |
|---|---|---|---|---|
| Iran | $A_1$ | 29.5 | 10.6 | 2.1 |
| | $A_2$ | 22.1 | 7.95 | 1.55 |
| | $A_3$ | 259 | 93.4 | 18.2 |
| | $A_4$ | 76.2 | 27.5 | 5.35 |
| | $A_5$ | 245 | 88.5 | 17.2 |
| | $A_6$ | 46.9 | 16.9 | 3.29 |
| | $A_7$ | 19.6 | 7.1 | 1.37 |
| | $A_8$ | 13.2 | 4.75 | 0.93 |
| | $A_9$ | 13.3 | 4.78 | 0.93 |
| | $A_{10}$ | 13.2 | 4.75 | 0.93 |
| Germany | $B_1$ | 36.3 | 13.1 | 2.55 |
| | $B_2$ | 10.7 | 3.85 | 2.55 |
| | $B_3$ | 9.95 | 3.59 | 0.75 |
| | $B_4$ | 17 | 6.15 | 0.7 |
| | $B_5$ | 57.9 | 20.9 | 1.2 |

## 3.6. Health risk assessment of PAEs

Consumption of tea prepared via teabag strongly affects the migration of PAEs compounds to the human body as determined in the present study. The HQ and CR levels of 6 PAEs congeners via tea consumption are shown in Table 4 and the EDI values of PAEs compounds are displayed in Table S6. As data on the long-term toxicology of DiBP accumulation is limited, the HQ value for DiBP was not estimated in this study. The HQ values of PAEs was as follows: DEHP > DOP > DMP > DBP > BBP > DEP. The HQ values of DEHP for 3 age groups (in the case of $A_2$ and $A_9$ brands), for children and teens (in the case of $A_1$, $A_7$, $B_1$, and $B_5$ brands), and for children (in the case of $A_4$, $A_5$, $A_8$, and $A_{10}$ brands) as well as DOP for $A_2$ brand were above 1, which indicates high risk for human health. Based on Table 4, the highest and lowest estimated HQ values for the Persian and German teabags were related to $A_2$ and $B_3$ brands, respectively. The findings of this research were in contrast with the findings of a former research carried out in Spain, which suggests that no PAEs compounds pose risk to human health (Alnaimat et al., 2020). Also, in a study in Italy, no PAEs congeners in tea infusions showed a HQ greater than 1 (Troisi et al., 2019). As presented in Table 4, three age groups are at risk of cancer due to DEHP intake via drinking tea prepared by teabags. Up to now, the carcinogenic risk of PAEs has not been evaluated for tea consumption to compare our results. Similar findings were observed in a study in Turkey through the consumption of coffee (Isci et al., 2023). According to the findings of this study, the teabags usage can cause harmful effects on human health considering the levels of PAEs.

Table 4
Risk assessment of 6 PAEs compounds in the different teabag brands for 3 age groups. The values with high risk are shown in bold fon

| PAEs compounds | Age groups | Teabag brand | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | $A_1$ | $A_2$ | $A_3$ | $A_4$ | $A_5$ | $A_6$ | $A_7$ | $A_8$ | $A_9$ | $A_{10}$ | $B_1$ | $B_2$ | $B_3$ |
| **Non-carcinogenic effects** | | | | | | | | | | | | | | |
| DMP | Children | 0.07 | 0.19 | 0.06 | 0.02 | 0.23 | 0.15 | 0.20 | 0.19 | 0.12 | 0.21 | 0.17 | 0.05 | 0.12 |
| | Teens | 0.02 | 0.07 | 0.02 | 0.01 | 0.08 | 0.05 | 0.07 | 0.07 | 0.04 | 0.08 | 0.06 | 0.02 | 0.04 |
| | Adults | 0.006 | 0.02 | 0.005 | 0.002 | 0.019 | 0.013 | 0.02 | 0.02 | 0.010 | 0.02 | 0.01 | 0.005 | 0.01 |
| DEP | Children | 8.61E-05 | 0.04 | 0 | 0 | 0.001 | 0 | 0.022 | 0 | 0 | 0.002 | 0.007 | 0.003 | 0.00 |
| | Teens | 3.1E-05 | 0.02 | 0 | 0 | 0.0004 | 6.69E-05 | 0.008 | 7.38E-05 | 8.03E-05 | 0.0006 | 0.002 | 0.001 | 0.00 |
| | Adults | 7.45E-06 | 0.004 | 0 | 0 | 0.0001 | 1.61E-05 | 0.002 | 1.77E-05 | 1.93E-05 | 0.0001 | 0.0006 | 0.0003 | 0.00 |
| DBP | Children | 0.15 | 0.04 | 0.003 | 0.01 | 0.04 | 0.03 | 0.05 | 0.06 | 0.01 | 0.03 | 0.15 | 0.04 | 0.00 |
| | Teens | 0.05 | 0.01 | 0.001 | 0.005 | 0.02 | 0.01 | 0.02 | 0.02 | 0.005 | 0.01 | 0.05 | 0.01 | 0.00 |
| | Adults | 0.01 | 0.003 | 0.0003 | 0.001 | 0.004 | 0.003 | 0.004 | 0.005 | 0.001 | 0.003 | 0.01 | 0.003 | 0.00 |
| BBP | Children | 0.001 | 0.02 | 0.0002 | 0.002 | 0.002 | 0.001 | 0.003 | 0.002 | 0.005 | 0.02 | 0.004 | 0.002 | 0.01 |
| | Teens | 0.0004 | 0.01 | 8.31E-05 | 0.0008 | 0.0006 | 0.0005 | 0.0009 | 0.0007 | 0.002 | 0.006 | 0.001 | 0.0006 | 0.00 |
| | Adults | 9.04E-05 | 0.002 | 1.99E-05 | 0.0002 | 0.0001 | 0.0001 | 0.0002 | 0.0002 | 0.0004 | 0.0015 | 0.0003 | 0.0001 | 0.00 |
| DEHP | Children | **6.76** | **20.75** | 0.55 | **1.63** | **2.67** | 0.69 | **2.77** | **1.17** | **12.94** | **1.71** | **3.08** | **1.96** | 0.78 |
| | Teens | **2.44** | **7.48** | 0.2 | 0.59 | 0.96 | 0.25 | **1** | 0.42 | **4.67** | 0.62 | **1.11** | 0.71 | 0.28 |
| | Adults | 0.58 | **1.8** | 0.05 | 0.14 | 0.23 | 0.06 | 0.24 | 0.1 | **1.12** | 0.15 | 0.27 | 0.17 | 0.07 |
| DOP | Children | 0.08 | **27.24** | 0.13 | 0.19 | 0.16 | 0.02 | 0.34 | 0.19 | 0.17 | 0.10 | 0.14 | 0.09 | 0.06 |
| | Teens | 0.03 | **9.83** | 0.05 | 0.07 | 0.06 | 0.006 | 0.12 | 0.07 | 0.06 | 0.04 | 0.05 | 0.03 | 0.02 |
| | Adults | 0.007 | **2.36** | 0.01 | 0.02 | 0.014 | 0.002 | 0.03 | 0.02 | 0.01 | 0.009 | 0.01 | 0.008 | 0.00 |
| **Carcinogenic effects** | | | | | | | | | | | | | | |
| BBP | Children | 3.97E-07 | 8.12E-06 | 8.75E-08 | 8.94E-07 | 6.21E-07 | 4.98E-07 | 9.82E-07 | 7.13E-07 | 1.88E-06 | 6.75E-06 | 1.36E-06 | 6.28E-07 | 4.04 06 |
| | Teens | 1.43E-07 | 2.93E-06 | 3.16E-08 | 3.22E-07 | 2.24E-07 | 1.8E-07 | 3.54E-07 | 2.57E-07 | 6.78E-07 | 2.44E-06 | 4.92E-07 | 2.26E-07 | 1.46 06 |
| | adults | 3.43E-08 | 7.03E-07 | 7.57E-09 | 7.73E-08 | 5.37E-08 | 4.31E-08 | 8.5E-08 | 6.17E-08 | 1.63E-07 | 5.85E-07 | 1.18E-07 | 5.44E-08 | 3.49 07 |
| DEHP | Children | 0.002 | 0.006 | **0.0002** | **0.0005** | **0.0007** | **0.0002** | **0.0008** | **0.0003** | **0.0036** | **0.0005** | **0.0009** | **0.0005** | **0.00** |
| | Teens | **0.0007** | 0.002 | **0.0001** | **0.0002** | **0.0003** | **0.0001** | **0.0003** | **0.0001** | **0.0013** | **0.0002** | **0.0003** | **0.0002** | **0.00** |
| | adults | **0.0002** | **0.0005** | **1.34E-05** | **3.96E-05** | **6.48E-05** | **1.66E-05** | **6.73E-05** | 2.84E-05 | 0.0003 | **4.13E-05** | **7.46E-05** | **4.75E-05** | **1.9E-05** |

## 4. Conclusion

In this work, the levels of MPs and PAEs in tea infusions prepared by different brands of Persian and German teabags were investigated. A size category of 100–250 μm was the most detected size of MPs, and the dominant shape of MPs was fibers. Furthermore, PE and nylon were the dominant types of polymer in the teabags. High PAEs concentrations in the Persian and German teabag samples were detected with average levels of 2.87 and 2.37 mg/g (4.76 and 3.93 mg/L), respectively. The higest levels of PAEs compounds in the teabags were related to DEPH and DiBP. According to the EDI levels of MPs from the tea consumption prepared by teabags for different age groups, the most exposure risk was specified for children. The health risk assessment of PAEs for teabags for different age groups showed that DEHP (in the Persian and German brands) and DOP (in the Persian brand) displayed great risks for children and teens. Also, the results showed that three age groups are at risk of cancer due to DEHP intake via drinking tea infusions prepared with teabags. According to the findings of this work, it can be concluded that the teabags consumption is an important human exposure route to MPs and PAEs. Knowledge of the levels and health effects of MPs and PAEs via tea consumption prepared by teabags is currently limited. Thus, further studies are required to demonstrate the mechanisms of MPs and PAEs release from teabags into tea infusions, the effective factors on the release of mentioned contaminants, and the contaminants that may be connected to MPs in the teabags.

# Declarations


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### Ethical Approval

Not applicable

### Consent to participate

Not applicable

### Consent for publication

Not applicable

### Author Contributions

Fatemeh Sadat Kashfi: Writing-original draft, Software, Analysis; Azam Mohammadi: Writing-original draft, Software, Analysis; Farzaneh Rostami: Sampling, Samples preparation; Arezou Savari: Sampling, Samples preparation; Gabriel E. De-la-Torre: Methodology, Writing-review & editing; Jörg Spitz: Conceptualization, Methodology; Reza Saeedi: Consulting, Writing-review & editing; Mohammadreza Kalantarhormozi: Consulting, Writing-review & editing; Akram Farhadi: Consulting, Writing-review & editing; Sina Dobaradaran: Supervision, Conceptualization, Methodology, Project administration, Final writing polishing.


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### Competing Interests

Authors have no conflict of interests.

### Availability of data

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

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## Figures

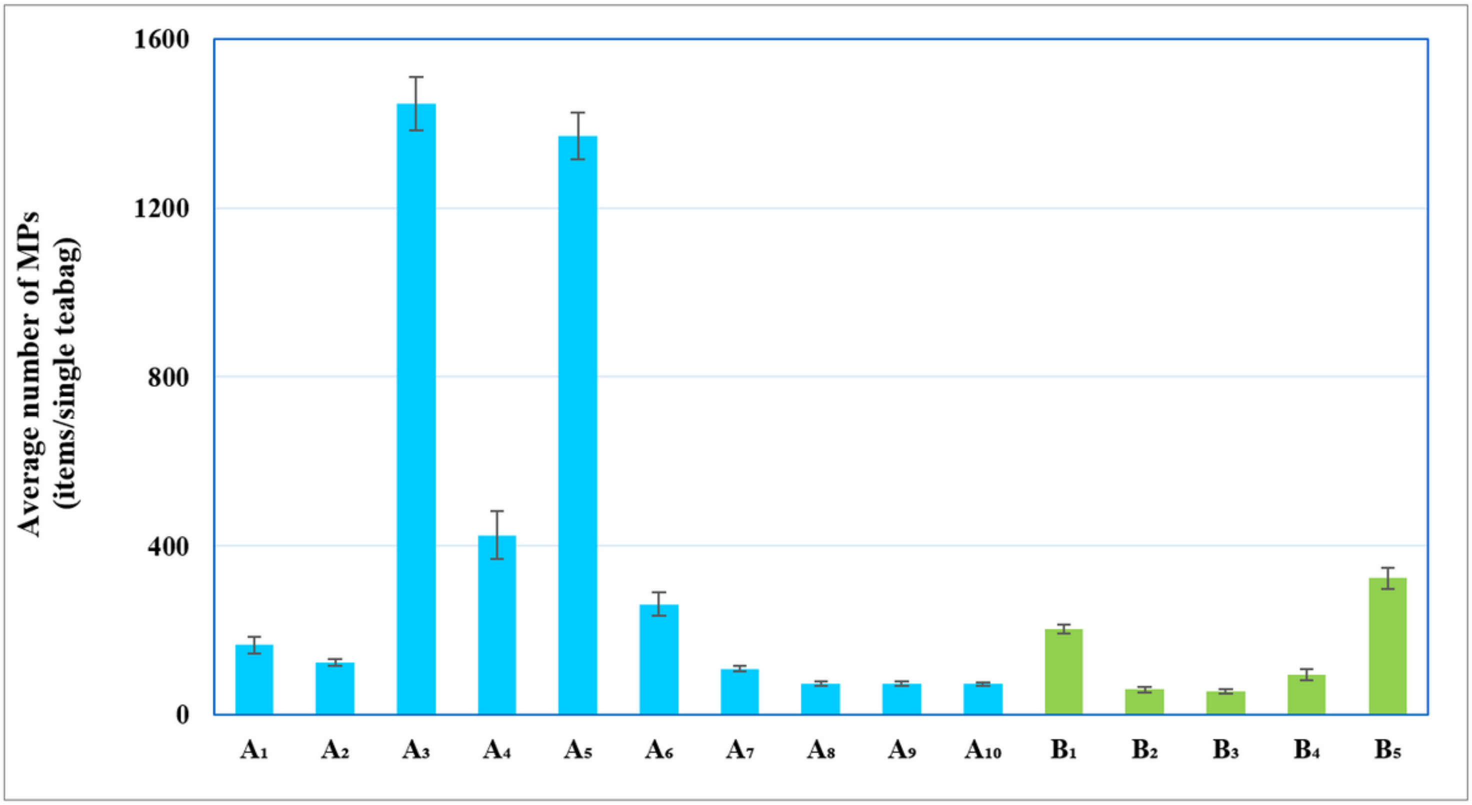



Figure 1

The average number of identified MPs in different brands of the Persian ($A_1$-$A_{10}$) and German ($B_1$-$B_5$) teabags.

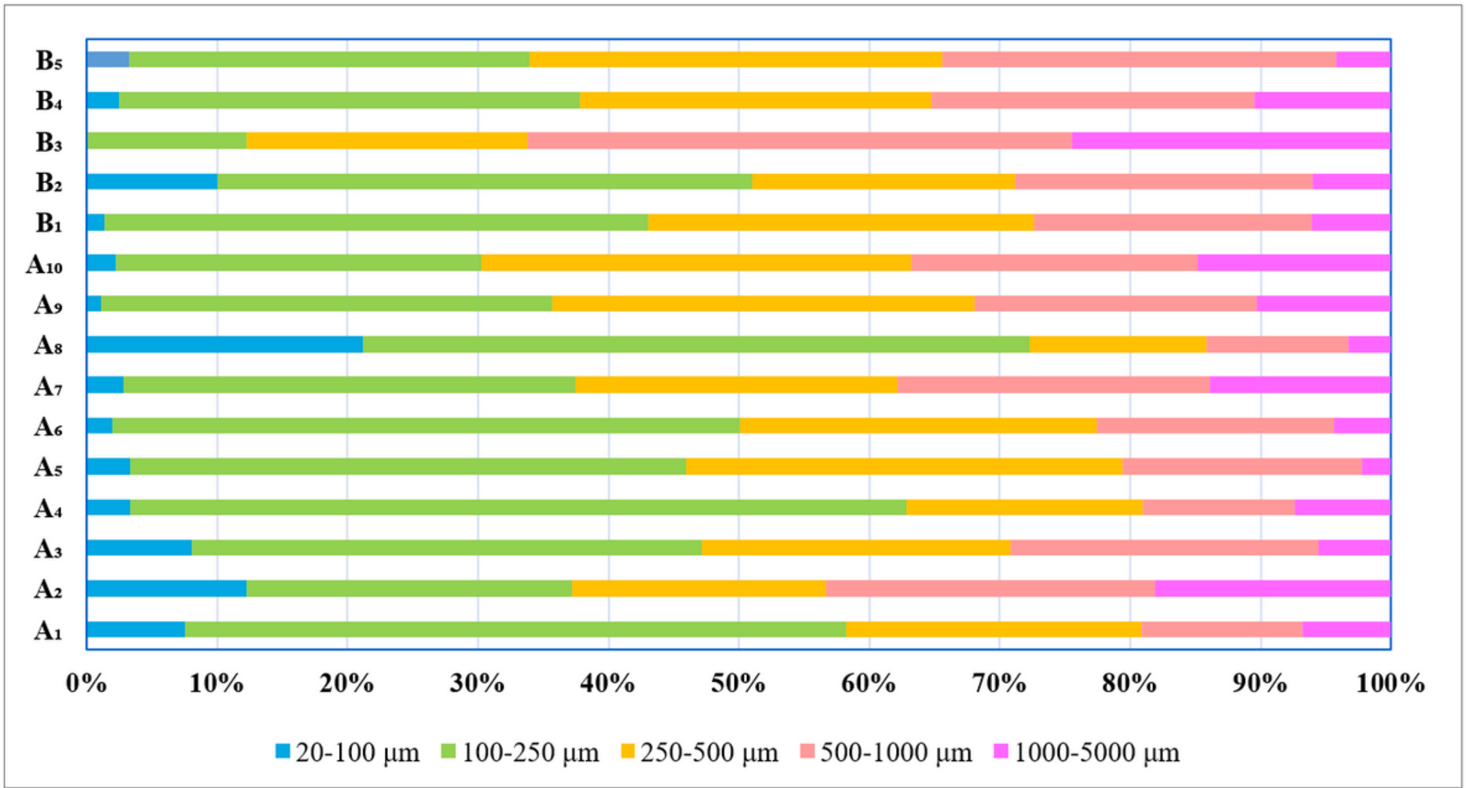
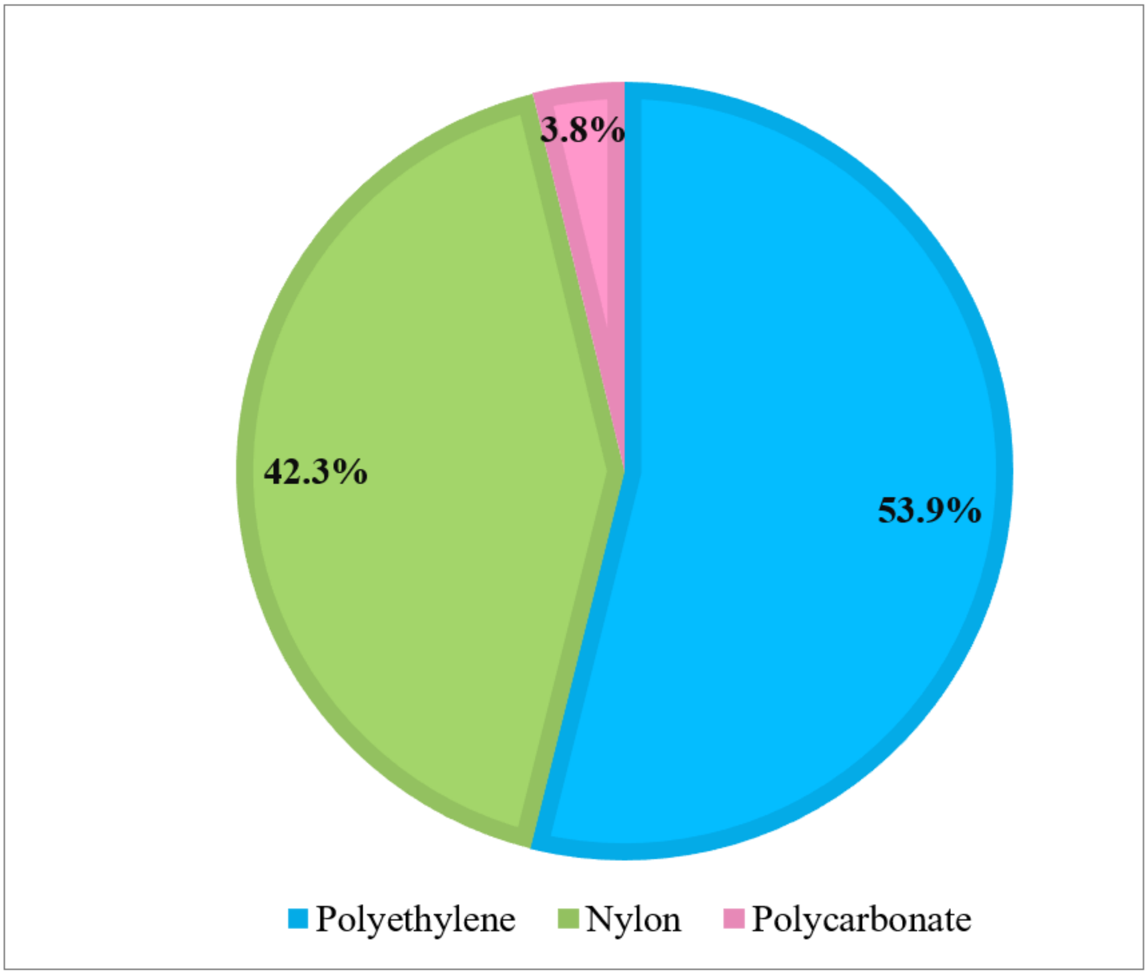


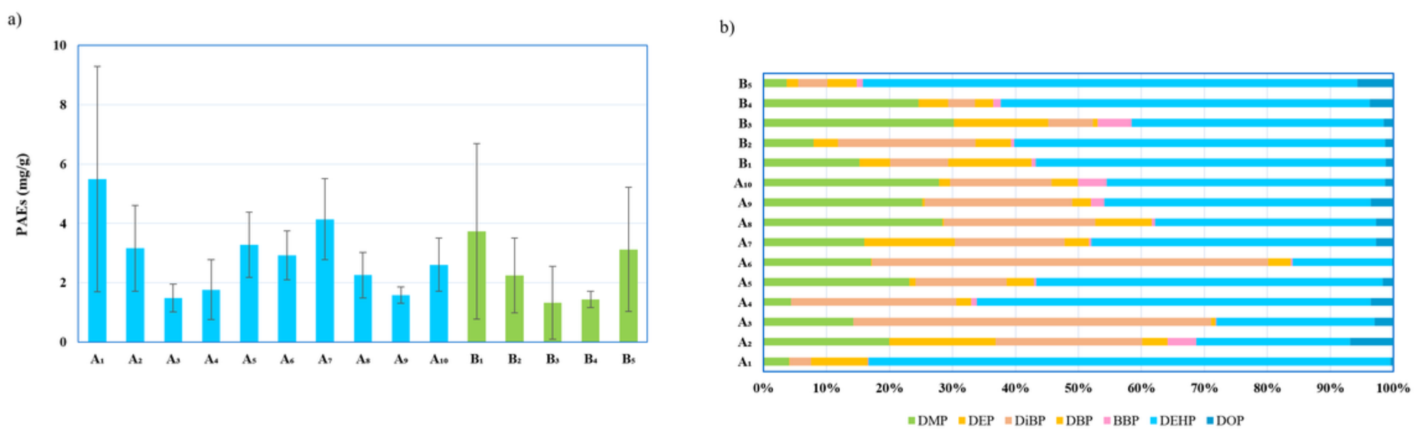

Figure 2

Size distribution of identified MPs in different brands of Persian ($A_1$-$A_{10}$) and German ($B_1$-$B_5$) teabags.




### Figure 3

Polymer type of detected microplastics in the teabag samples.




### Figure 4

Total PAEs concentrations (a) and percentages of detected PAEs compounds (b) in different brands of the Persian ($A_1$-$A_{10}$) and German ($B_1$-$B_5$) teabags.

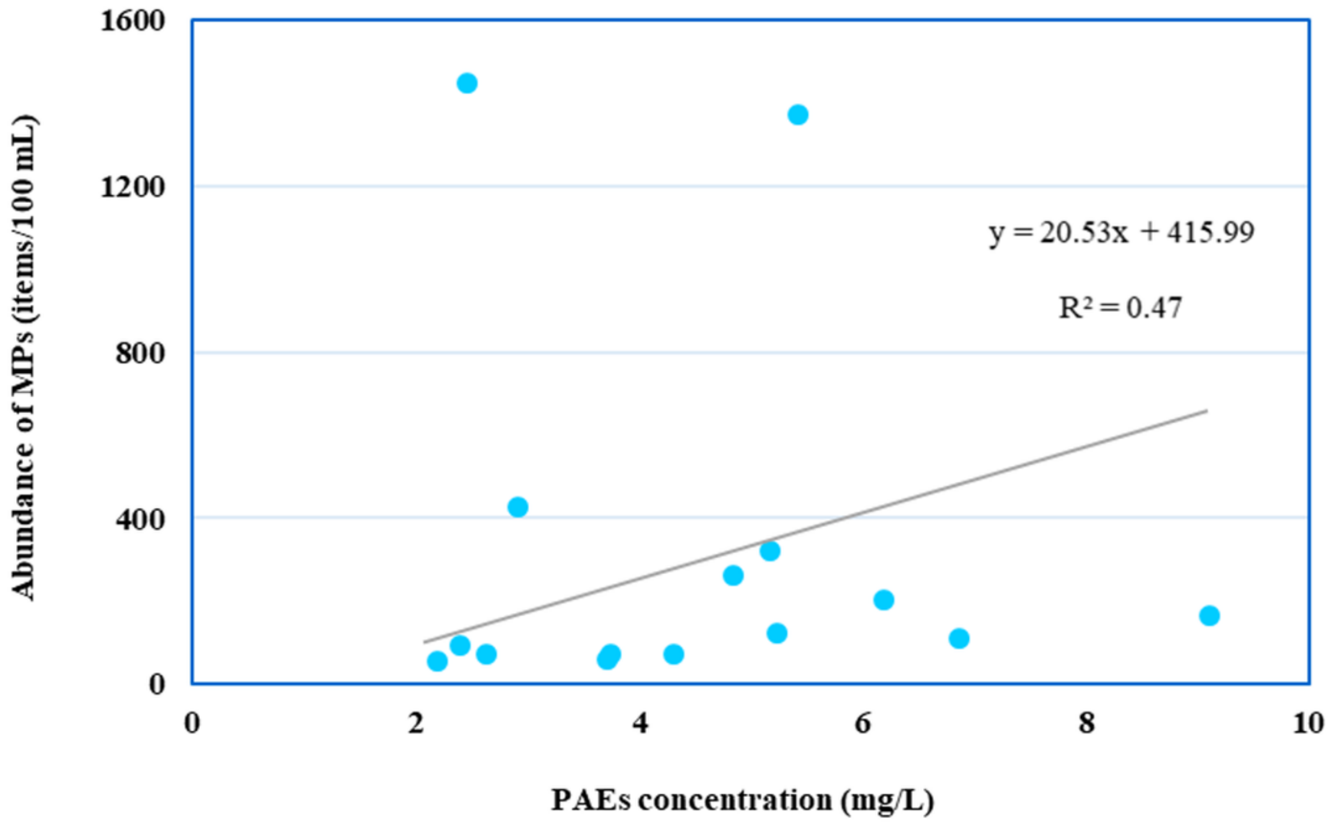



**Figure 5**

Correlation between MPs abundance and PAEs concentration leachates via teabags.

## Supplementary Files

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