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Cigarette Filters as Long-Term Vectors of Microfibers in Marine Environments- a Case Study

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Abstract

Introduction: Introduction: Cigarette filters (CFs) are a potential source of microfibers (MFs) in aquatic environments. In this study, MFs released from two CFs brands into seawater (SW) and distilled water (DW) under outdoor and laboratory environmental conditions during various long-term exposure times (120, 180, 240, and 365 days) were investigated and compared.

Methods: Unsmoked CFs from two commercially available cigarette brands were exposed to SW and DW under outdoor and laboratory conditions at contact times of 120, 180, 240, and 365 days. Three CFs of each brand were used for each contact time under every environmental condition. Determining the characteristics of MF were performed microscope and Raman techniques.

Results: Average MF levels released from CFs into SW and DW increased over time, from 120 to 365 days, from 41.6 to 61.4 and 24.0 to 46.9 items/g CFs, respectively. The 20-50 µm size range was the most abundant. Cellulose acetate (90%) and polypropylene (10%) were the identified polymer compositions.

Conclusion: These findings highlight that CFs play an important role in MFs release into the environment. Thus, there is an urgent need to develop strategies for reducing cigarette waste in the environment.

Keywords: Aquatic, Environment, Microplastics, Smoke, Tobacco products

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Introduction

Cigarette butts (CBs) are among the most frequently discarded forms of litter worldwide. It is estimated that 967 million smokers worldwide consume approximately 6 trillion cigarettes annually, of which 4.5 trillion CBs are discarded into the environment. Improper disposal of CBs is a common problem, as smokers often incorrectly dispose of them into the environment. Cigarettes are composed of a cellulose acetate filter, tobacco, and wrapping paper. Cigarette filters (CFs) may contain functional additives like plasticizers, flame retardants, and softeners (1). CFs discharged into the environment may be transmitted to the marine ecosystems in different ways. The littered CFs contain various toxic compounds, leached into the surrounding environment and cause serious ecological risks (2). Microfibers (MFs) are the predominant form of microplastics (MPs) released from CFs. The detection of MFs released from CFs is of particular environmental relevance because fibers have also been reported as a predominant MP morphology in aquatic ecosystems, including the Persian Gulf region (3). After entering marine environments, CFs undergo physical abrasion, photodegradation, and chemical weathering, leading to the fragmentation and release of MFs (4), which may be consumed mistakenly by aquatic species and cause harmful effects.

The lack of information on prolonged MF release from CFs represents an important knowledge gap because

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discarded CFs may remain in the environment for extended periods. Understanding whether MF release continues or changes during long-term exposure is necessary for assessing the potential contribution of cigarette waste to persistent MF contamination. In addition, comparison of different water types and environmental conditions can provide further information on whether MF release varies according to the surrounding aquatic environment. Because discarded CFs, as a potential source of MFs (4,5), plastic additives, and other toxic compounds (2) can cause ecological hazards, it is necessary to determine the role of CFs in releasing MFs into the environment. To this end, in phase I of this work, the levels and features of released MFs from CFs into different types of water at short exposure times of 1, 2, 7, 15, 30, and 60 days, ecological risk assessment, and the emission rate of MFs via CFs into aquatic environments were considered (4). However, the release of MFs from CFs under long exposure times remained insufficiently characterized. Thus, continuing phase I, this study (Phase II) aimed to investigate and compare the levels and features (size, color, and polymer type) of MFs released from CFs into seawater (SW) and distilled water (DW) during longer exposure times of 120, 180, 240, and 365 days. It should be noted that the comparison of SW and DW was conducted to determine whether water type influences the long-term release and features of MFs from CFs. The emission rate of MFs via CFs into marine environments on a worldwide scale was also considered.

Materials and Methods

Study design

The cigarette sampling and identification of MFs released from CF samples were done based on phase I of this work (4). In January 2023, a total of 96 cigarette samples of two commercially available international brands [as brand 1 (n = 48) and brand 2 (n = 48)] were collected from a market in Bushehr, Iran. Two cigarette brands were selected based on their commercial availability in the local market, and an equal number of samples from each brand was collected to ensure balanced representation in the experimental design. Experiments on MF release from unsmoked CFs were performed in two types of water (SW and DW) under two environmental conditions [outdoor environment (OE) and lab environment (LE)] at four contact times of 120, 180, 240, and 365 days. The exposure periods of 120, 180, 240, and 365 days were selected to extend the exposure duration investigated in Phase I (1-60 days) and to evaluate MF release during progressively longer environmental exposure. The selected times were intended to provide information on MF release over longer-term exposure periods. The average weight (± SD) of brand 1 and brand 2 CF samples was 0.175 (± 0.003) and 0.173 (± 0.003) g, respectively. SW and DW samples, obtained from the Persian Gulf and water purification system (Lane End, High Wycombe, UK), respectively, were used for experiments after filtering through polytetrafluoroethylene (PTFE) membrane filters (Whatman, Maidstone, Kent, UK) with a pore size of 0.45 μm.

Sample processing and microfiber identification

After cutting the CFs to a length of 3 cm using a sterile stainless-steel blade, each unsmoked CF sample of each brand was placed in a 250-mL glass beaker. The CF samples were exposed to four conditions: SW/in LE, SW/in OE, DW/in LE, and DW/in OE. 100 mL of water (SW or DW) was added to each beaker, which was then covered with aluminum foil, and maintained under static condition at different time periods of 120, 180, 240, and 365 days. Three CFs of each brand were separately placed in glass beakers for each contact time under every environmental condition. After the desired exposure times, each sample was vacuum-filtered through a PTFE membrane filter with a pore size of 0.45 μm. Then, filters were separately placed into clean petri dishes and air-dried at room temperature.

The dried filter papers were analyzed for the presence of MFs using an optical microscope (A. KRÜSS Optronic, Germany) with a maximum magnification of 40x to evaluate their size and color. To accurately distinguish MFs from other potential crystalline structures, such as salt particles and chemical residues, a heated needle test was performed when necessary. The longest dimension of each MF was measured using the diameter of a stainless-steel probe. Moreover, a DXR3 Raman Microscope (Thermo Fisher) with a 10x magnification lens was applied to identify the polymer type of selected MFs (40 particles). All MF samples were placed onto prepared copper tapes for analysis, and a 785 nm laser was applied as the excitation source. The Raman spectra were obtained in the range of 400-2450 cm^{-1}. The types of MFs were identified by the obtained spectra to a reference database library for the microscope. The polymer composition of MFs was identified by comparing the obtained Raman spectra with the reference spectra.

Quality assurance (QA)/quality control (QC)

To prevent contamination, cotton lab clothes and nitrile gloves were used in all experimental steps. The water samples were filtered before use and stored in glass bottles. All used glassware was rinsed with DW and methanol. Paper wipes were utilized to cleanse all desks. Four blank samples were also provided during each sample set. It is worth noting that 3, 4, 4, and 5 items were detected in two blank SW samples at exposure times of 120, 180, 240, and 365 days, respectively. Similarly, 2, 3, 4, and 3 particles were detected in two blank DW samples at corresponding contact times, respectively. The number of MFs measured in the blank samples was subtracted from the values of experimental samples to remove the error due to any potential contamination.

Statistical analysis

Microsoft Excel 2018 and SPSS 20 software were used for statistical analyses. Each individual CF was considered the independent experimental unit. Three independent filter replicates were analyzed for each combination of

cigarette brand, water type, environmental condition, and exposure time. MF release was expressed as items/g CFs for each individual filter. Shapiro-Wilks and Levene tests were applied to assess the normality and homogeneity of variances. The data were distributed normally, and the homogeneity assumption of variances was confirmed. The differences in the levels of MFs released from CFs at four contact times were investigated using one-way analysis of variance (ANOVA). Moreover, the differences in the levels of released MFs across cigarette brands, water types, and environmental conditions were analyzed using independent t-test analysis. A *P* value of less than 0.05 was considered statistically significant. The effect sizes for exposure time, water type, cigarette brand, and environmental conditions were also estimated using Cohen's d. Cohen's d of 0.2, 0.5, and 0.8 indicate small, medium, and large effects, respectively.

Results

Levels of microfibers

The number of identified MFs released through CFs into water at various exposure times is presented in Table S1. The levels of MFs released via CFs into SW and DW in LE and OE conditions at contact times of 120, 180, 240, and 365 days are also presented in Figure 1 and Table S2. The average (±SD) levels of released MFs into SW at exposure times of 120, 180, 240, and 365 days were 41.6 (±8.82), 49.0 (±10.3), 53.3 (±2.90), and 61.4 (±6.13) items/g CFs, respectively. Also, the average (±SD) MF levels in DW during the studied contact times were 24.0 (±4.69), 27.0 (±4.2), 33.1 (±6.44), and 46.9 (±8.79) items/g CFs, respectively (Figure 1 and Table S2).

Characteristics of MFs

Figure S1 (a) and Table S3 show the distribution of MFs released via CFs into SW and DW at the studied exposure times based on their size range. MFs with a size range of 20-50 µm were the most abundant, followed by size ranges of 50-100 µm, 100-250 µm, and 250-500 µm. As depicted in Figure S1 (b) and Table S3, the detected MF colors were white/transparent, yellow/brown, and black/gray. White/transparent was the dominant color in both water types and different exposure times. The identified polymer compositions of MFs were cellulose acetate (90%) and polypropylene (10%), as shown in Figure S2. The Raman spectra of some found polymer compositions are also displayed in Figure S3.

Emission rate of MFs

The global release of MFs into the water environment was evaluated based on the average levels of MFs released from CFs into SW at the studied exposure times and ~845000 tons of discarded CFs each year worldwide (6). The potential global MF emission was calculated using the following equation:

$$\text{Global MF emission}\left(\text{items / year}\right) = \textit{MF release rate}\left(\textit{items / g CFs}\right) \times \text{annual mass of discarded CFs}\left(\text{g / year}\right)$$

As seen in Figure 2 a and b, the emission rates of MFs via CFs into SW were estimated 7×10^{12}-22×10^{12} items during exposure times of 1-60 days (4) and 36×10^{12}-52×10^{12} at 120-365 days of exposure.

Discussion

Levels of microfibers

Based on Figure 1 and Table S2, the average (±SD) levels of MFs released into SW at exposure times of 120, 180, 240, and 365 days were 41.6 (±8.82), 49.0 (±10.3), 53.3 (±2.90), and 61.4 (±6.13) items/g CFs, respectively. The average (±SD) MF levels in DW during the studied contact times were also 24.0 (±4.69), 27.0 (±4.2), 33.1 (±6.44), and 46.9 (±8.79) items/g CFs, respectively. While in phase I of the study (4), the average levels of released MFs into SW and DW ranged from 8.21 to 26.1 and 1.91 to 14.2 items/g CFs,

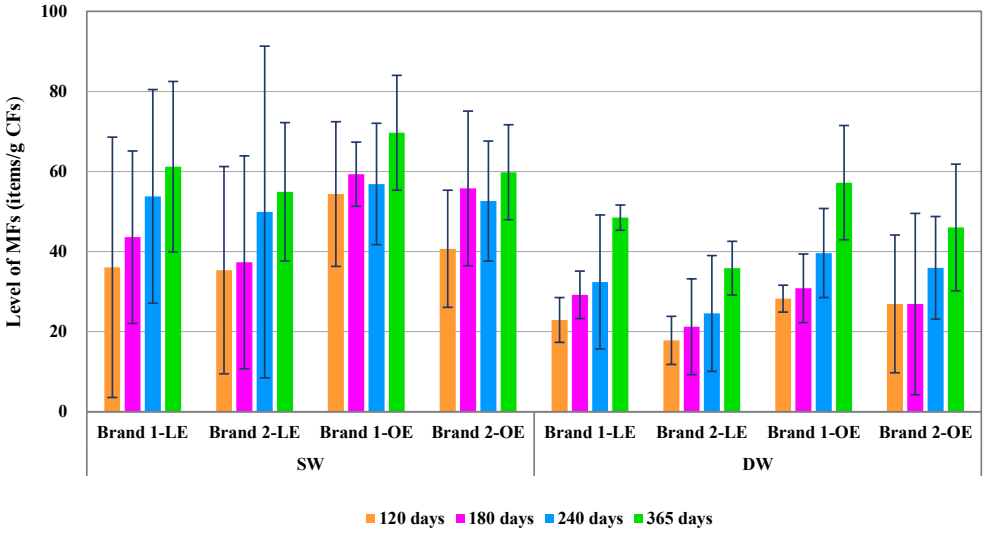


Figure 1. Microfibers (items/g CFs) levels released from cigarette filters into two water types at four exposure times (SW: seawater, DW: distilled water, LE: lab environment, and OE: outdoor environment). Values represent the average of three independent experimental replicates (n=3), and error bars represent (SD: standard deviation). MF counts were corrected by subtracting the corresponding blank value. The levels of MFs released from CFs were calculated by normalizing the particle count to the mass of the corresponding CF and expressed as items/g CFs.

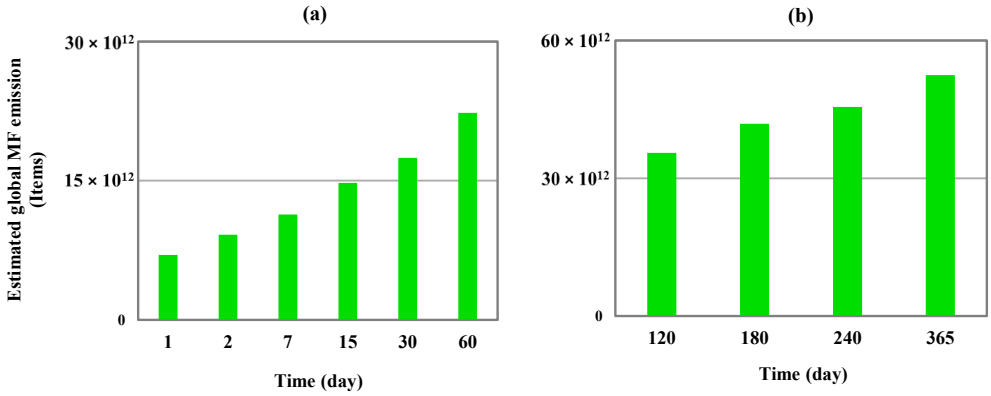


Figure 2. Emission rate of microfibers via cigarette filter into seawater at exposure times of 1-60 days (a) [data presented in phase I (4)] and 120-365 days (b)

respectively, during exposure times of 1 to 60 days. The progressive increase in MFs released from CFs indicates a time-dependent pattern of release. In phase I of this work, the release of MFs via CFs into water increased during 1 to 60 days, indicating that initial liberation primarily involves the detachment of surface fibers. Whereas prolonged exposure (120-365 days) weakens the CF structure and promotes greater MF release. These findings imply that CFs act as a continuous MF source, contributing to the accumulation of synthetic fibers in marine ecosystems.

In a previous study in Terrassa, Spain, the MF levels released from smoked CFs via various brands of cigarettes were investigated in the lab environment under continuous exposure to UV light and slow mechanical agitation for one month. It was reported that 100 MFs per day, less than 0.2 mm in size, were released from each smoked CF during the initial two weeks (5). As illustrated in Table S2, a statistically significant difference was found in the levels of released MFs among the various exposure times (P value < 0.001 and effect size = 2.16) and two types of water (P value < 0.001 and effect size = 1.76). The levels of MFs released from CFs were not significantly different among the various cigarette brands (P value = 0.56 and effect size = 0.63) and environmental conditions (P value = 0.60 and effect size = 0.48). It should be noted that these findings are consistent with phase I of this study (4). In contrast, the release rates of MFs from surgical face masks into aquatic environments, as reported in a study in Bangladesh, were significantly influenced by environmental parameters such as water salinity or turbulence (7). The present study used prolonged exposure periods of 120-365 days, during which the time-dependent release of MFs may have been more influential than differences between the two environmental conditions. Also, the samples under OE condition were exposed to natural environmental conditions, while temperature and UV intensity were not continuously monitored or quantified. Therefore, the differences in temperature and UV exposure between OE and LE conditions could not be determined.

The release of MFs, especially cellulose acetate, from CFs into seawater is influenced by the interaction of environmental, chemical, and physical factors. The duration of exposure is one of the most important determinants of MF release. The number of MFs released from CFs is expected to increase progressively over time due to photooxidation, mechanical degradation, and hydrolysis (5). High salinity and low alkalinity, typical of marine waters, can promote ionic interactions between salt ions and the acetyl groups of cellulose acetate, thereby facilitating structural loosening and accelerating fiber detachment (8). This may explain the higher release of MFs via CFs into SW observed in this study. Water turbulence also influences the release of MFs from CFs (9), which was not tested in the present study. In addition to MFs, CFs leach different chemicals into aquatic environments. Therefore, CFs act not only as a source of MFs but also as a vector of chemical contaminants. The released MFs and their chemical leachates can bioaccumulate in the organisms, potentially influencing the food webs and ecosystem (2).

Characteristics of MFs

As seen in Figure S1 (a) and Table S3, MFs with a size range of 20-50 µm were the most abundant at the studied exposure times. As reported in phase I (1-60 days) (4), larger size ranges (250-500 µm and 100-250 µm) dominated at early exposure times (1, 2, and 7 days). While at other exposure times (15, 30, and 60 days), the size range of 20-50 µm was dominant. The results of phases I and II showed that prolonged environmental weathering and polymer aging can progressively fragment MFs into smaller particles. The observed size distribution pattern highlights a multi-stage degradation pathway, starting with the release of larger MFs and continuing with the accumulation of fine MFs. These results underscore the role of CFs as a long-term and continuous source of MF contamination in marine ecosystems. Larger plastic particles can break down into smaller particles over time due to UV radiation, mechanical abrasion, and microbial colonization (10). Similar findings have been reported for textile-derived MFs in an Italian study. As reported, initial MFs released from synthetic textiles often featured larger (> 100 µm) fibers, which subsequently fragmented into smaller fibers (< 50 µm) after repeated washing and environmental exposure (11). Additionally, this study's results were consistent with findings from a previous Swedish study that plastic debris fragmented into smaller particles with increasing exposure time (12). From an environmental risk perspective, MP

particles smaller than 50 μm are very important. Smaller MPs have greater potential for ingestion by lower trophic-level organisms, such as zooplankton, and are more easily translocated across biological membranes. Moreover, a higher surface area to volume ratio of MPs can enhance their capacity to adsorb chemical contaminants, increasing their potential ecotoxicological impacts (13).

The identified MF colors were white/transparent, yellow/brown, and black/gray, based on Figure S1 (b) and Table S3. Similar to phase I of this study (4), white/transparent was the dominant color in both water types and different exposure times. The dominance of white/transparent MFs released from CFs can be attributed to the color of cellulose acetate fibers used in CF manufacturing (14). Yellowing of polymers in phase II can occur due to UV radiation over extended timescales, and this discoloration may be limited to the surface layers (10). In parallel, fibers in aquatic environments are prone to adsorb dissolved organic matter, humic substances, and metal ions, further darkening MFs (15). The variety of MP colors reflects the diversity of their contamination sources in the environment (16). Due to the resemblance of MPs to prey, organisms may mistakenly ingest them, which may lead to digestive blockages, internal injuries, false signals of satiety, nutritional deficiencies, and even introduction of harmful chemicals and pathogens into their bodies (17).

As illustrated in Figure S2, the detected polymer types of MFs were cellulose acetate (90%) and polypropylene (10%). Similar to phase I (4), this study confirmed that cellulose acetate is the predominant polymer type in CFs, consistent with prior studies (5). The dominance of cellulose acetate in both experimental phases indicates that CFs are a major and long-term source of cellulose acetate in the coastal and marine ecosystems (18). Polypropylene is used as an additive in cellulose acetate (19). The shift from polyamide and polyethylene in phase I to polypropylene in phase II may be due to environmental weathering, including UV degradation and deacetylation of cellulose acetate. Deacetylation increases the surface area and degradation of cellulose acetate, possibly enabling the release of additives or secondary polymers (20). Although environmental exposure may influence the physical characteristics and degradation of materials, the present study did not directly investigate polymer transformation or establish a mechanism by which polyamide or polyethylene could produce polypropylene. Also, this result may be attributed to the fragmentation of polyethylene and polyamide fibers into smaller particles (< 20 μm) that were not detected in the present study, as the main limitation of this work.

Emission rate of MFs

The emission rates of MFs through CFs into SW increased from 7×10^{12}-22×10^{12} items within 1-60 days of exposure (4) to 36×10^{12}-52×10^{12} during an exposure time of 120-365 days, as demonstrated in Figure 2 a and b. However, it is worth noting again that MFs smaller than 20 μm were not identified in the current study, so the emission rates reported here are likely underestimated. Therefore, the actual emission of MFs released via CFs is presumably much higher. The progressive increase in total emissions over time reflects the combined effects of photodegradation, hydrolysis, mechanical abrasion, and biofilm activity, which progressively weaken the filter structure and promote fragmentation into microfibers (21). This estimated MF emission rate highlights the potential of CFs to release trillions of MFs into aquatic ecosystems annually. These findings underscore that CFs are a potentially significant source of MFs entering aquatic environments. Because the global estimate of MF emissions is based on CFs from only two cigarette brands and on release rates obtained under specific environmental conditions, it should be regarded as a high-order potential estimate with substantial uncertainty. It does not represent a direct measurement of the actual global MF flux from discarded CFs into aquatic environments. Given the environmental burden of released MFs, their persistence, physical ingestion by living organisms, and interaction with adsorbed contaminants, the CBs waste requires more attention.

Limitations

Several limitations must be investigated when interpreting the results of the present study. The absence of standardized methods for the identification and quantification of MPs reduces the comparability of results among different studies. Conventional approaches for MF quantification generally focus on particle number rather than mass. In the present study, only MFs larger than 20 μm were identified through visual observation. This represents a major limitation because smaller MFs (< 20 μm) could not be detected, potentially resulting in a substantial underestimation of the total number of MFs released from CFs.

Another limitation was the characterization of MF polymer types. Due to the large number of suspected MF particles detected, not all particles could be analyzed using micro-Raman spectroscopy. Consequently, some uncertainty may remain regarding the polymer composition of the identified MFs. An important limitation of this study is that environmental variables such as temperature, UV radiation, hydrodynamic turbulence, mechanical abrasion, sediment presence, and biofilm formation were not incorporated into the experimental design, which should be investigated in future studies. Also, the small sample size as a limitation of present study that limits the statistical power of the comparisons and increases the possibility of type II error. Therefore, non-significant differences should not be interpreted as definitive evidence that a particular factor has no effect on MF release. These limitations should be taken into investigated in future research.

Conclusion

The results of this study clearly showed that the release of MFs from CFs increases in the aquatic environment over time. The levels of MFs released into SW were higher compared with DW. Also, the most common size range and color of MFs were 20-50 μm and white/transparent,

respectively. Cellulose acetate and polypropylene were the identified polymer compositions of MFs. On a global scale, the potential contribution of MFs derived from CF is substantial, with a huge number of fibers potentially entering aquatic ecosystems annually. This study provides clear evidence that CFs are a significant source of MFs in marine environments. Thus, further studies using larger sample sizes as well as advanced analytical techniques to identify smaller MPs (< 20 μm) are required. Given that cigarette waste is a source of various contaminants, implementing effective strategies to reduce this waste is needed.

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Competing Interests
The author declares that there is no conflict of interests.

Ethical Approval
This study was approved by the Research Ethics Committee of the Bushehr University of Medical Sciences (Ethical code: IR.BPUMS.REC.1402.310). The guidelines for conducting the research, as outlined by the committee, were strictly followed.

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Supplementary Files
Supplementary file 1 contains Figure S1-S3 and Tables S1-S3

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