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Research on the presence of cigarette butts and their leaching of chemical pollutants and microparticles: the case of Dalian, China

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Cigarette butts (CBs) can contain high amounts of toxic substances such as heavy metals, polycyclic aromatic hydrocarbons (PAHs) and microplastics, all of them can result from the incomplete decomposition of CBs. However, limited research exists on the pollution caused by littered CBs in China and the emissions of toxic substances from different Chinese cigarette brands.Therefore, the main objective of this study was to assess the quantity of CBs littered in Dalian city, China, and to evaluate the emissions of heavy metals, PAHs, and microplastics from different cigarette brands. During the survey, a total of 10,591 CBs items and 35 cigarette brands were collected. ICP-MS analysis confirmed the presence of 17 types of heavy metals in the highest abundance brand of CBs, with concentrations ranging from 0.02 μg/L (U and La) to 395 μg/L (Mn). In the lowest abundance brand of CBs, 15 types of heavy metals were detected, with concentrations ranging from 0.08 μg/L (U) to 297 μg/L (Mn). Additionally, a total of 9 PAHs, including compounds such as naphthalene and acenaphthylene, were detected in both the highest and lowest abundance brands of CBs using HPLC-MS/MS, with concentrations ranging from 0.40 ng/L (Anthracene) to 92.12 ng/L (Naphthalence). Microplastics and particles were also identified by light microscopy and SEM. Based on cigarette consumption data in China, it is estimated that annually 360,000 tons of plastic waste are generated by CBs in the environment. Considering the presence of CBs in urban and beach environments, the release of these toxic substances poses a threat to water resources.

KEYWORDS

cigarette butts, density, leachate, heavy metals, microplastics, PAHs

1 Introduction

Globally in 2019, it was estimated that about 1.13 billion smokers consumed 7.41 trillion tobacco products, and up to 75% of smokers were reported to throw their cigarette butts (CBs) into the environment at will (Rahman et al., 2020; Collaborators, G. B. D. Tobacco 2021). These discarded CBs have been found in various public sites such as parks, highways, and coastlines. They can also be transported from land into watercourses and oceans by rain, river flows, and floods, causing severe environmental risks on a widespread scale (Novotny et al., 2009; Ribeiro et al., 2022). Although CBs have been recognized as a widespread form of solid waste worldwide, accounting for approximately 38% of the total discarded waste according to environmental researchers, the public has a poor understanding of the environmental risks associated with discarded CBs (Curtis et al., 2017; Qamar et al., 2020; Soleimani et al., 2023b).

CBs were designed to reduce the harm of smoking, as they are generally composed by microfiber bundles of cellulose acetate which were believed to reduce inhaled toxic (Pauly et al., 2002; Novotny et al., 2009). Nevertheless, CBs not only fail to offer any health benefits but also increase the health risks as fibers are breathed in by smokers (Conradi and Sanchez-Moyano, 2022). Additionally, CBs are made from acetic acid-treated plant-derived cellulose, they are categorized as a bioplastic (Cheng et al., 2010). However, there is no evidence to show that cellulose acetate undergoes complete degradation under normal natural conditions (Bonanomi et al., 2020). Based on published data, about 20% to 30% of the mass of CBs will be lost for 2 years (Bonanomi et al., 2020). When CBs break down, microplastics and nanoplastics can persist in the environment, which can be harmful to soil and aquatic ecosystems (Chevalier et al., 2018; Belzagui et al., 2021; Dobaradaran et al., 2021b; Soleimani et al., 2023a). Therefore, CBs pose a double problem, as they not only fail to protect the health of smokers but also contribute to the accumulation of non-biodegradable waste in the environment.

Evidence has demonstrated that nearly 100 microplastic fibers were released from CBs per day with an average length less than 0.22mm and about 300,000 tones of potential microplastic fibers entered aquatic environment per year (Belzagui et al., 2021; Yang S. et al., 2023). These abundance released microplastic fiber have present an ecotoxicity to aquatic lifeforms such as fishes, molluscs and copepods (Belzagui et al., 2021; Conradi and Sanchez-Moyano, 2022). Meanwhile, CBs can release approximately 7000 chemical substances, about 200 of which are highly toxic and have the potential to be mutagenic or carcinogenic (Slaughter et al., 2011; Lima et al., 2021; Soleimani et al., 2022). Some of these substances are heavy metal, benzene, xylene, BTEX, PAHs, AAs and toluene. (Dobaradaran et al., 2018b; Dobaradaran et al., 2020; Akhbarizadeh et al., 2021a; Dobaradaran et al., 2021a; Dobaradaran et al., 2022; Dobaradaran et al., 2023; Soleimani et al., 2023b). Therefore, inappropriately disposed CBs caused a unique challenge to ecosystems.

Considering the environmental hazards caused by discarded CBs and the lack of information on the environmental risks associated with CBs littering in Chinese cities, it is crucial to conduct a survey to assess the presence of CBs and the potential leakage of hazardous substances into the environment. This survey will help raise awareness among the Chinese population about the issue and the data will be essential for the development of effective environmental protection strategies. Surveys took place in Dalian, a metropolis of Liaoning province of China, with approximately 6.1 million people as of 2022 and 2211km of coastline. The presence of CBs was evaluated in different of settings including Blackrock Reef beach, Xinghai park and Shuangxing City big vegetable market. Our objectives encompassed quantifying the extent of CBs littering in diverse environments and estimating the potential leakage of hazardous substances into the environment resulting from CB littering.

2 Materials and methods

2.1 Site description

Dalian is located on the southern part of the Liaodong Peninsula in Liaoning Province, bordering the Yellow Sea and the Bohai Sea, and is backed by the Northeast China hinterland while facing the Shandong Peninsula across the sea. It is renowned not only for its importance in ports, trade, commerce, and tourism but also for being one of the most livable cities in China (Zhang et al., 2021). In this study, the three sidewalks in Blackrock Reef beach, Xinghai park and Shuangxing City big vegetable market were selected for the CBs littering observation (see Supplementary Figure 1). These three areas represent typical human activity zones, characterized by beach and urban environments.

2.2 Data collection

Sampling took place in June 2023, a month within the peak tourist season in Dalian, which typically spans from May to September. The survey routes were chosen randomly at the three sampling areas were 2 km in length and 3 m in width for the park area, 2.57 km in length and 4 m in width for the beach area, and 3.77 km in length and 3 m in width for the market area (see Supplementary Figure 1). The CBs were collected in garbage bags and later sorted and counted in the laboratory. The daily collection occurred from 4:00 PM to 11:00 PM, as this time period follows the morning cleaning activities and approaches the evening, approximately representing the daily generation of CBs waste. All sampling campaigns were performed in duplicate over different days of the week.

2.3 Analysis of microplastics, heavy metals, and PAHs in CBs leachate

In order to analyze the leaching substances of CBs, we selected cigarette brands of CBs with the highest and lowest abundance. The cigarettes were systematically smoked using a pump under identical conditions (see Supplementary Figure 2). For each brand, 20 CBs were combined with 1L of deionized water and gently stirred to simulate disturbance in a water environment. After 12 hours, the leachate was first filtered three times with a 125 μm mesh filter and then subjected to vacuum filtration using a vacuum pump (Jin Teng, GM-0.33A, Tianjin, China) through a 0.22 μm polycarbonate (PC) membrane (Merck Millipore, Ireland). Once the filtration was complete, the membranes were transferred to glass petri dishes and dried in a drying cabinet at a temperature of 50°C for 2 hours. Procedural blanks were also conducted for each batch, which involved filtering 1L of deionized water solely through the membranes and subsequently drying them for 2 hours at 50°C. The samples on the membranes were used for microplastic detection, while the filtered liquid was used for the analysis of heavy metals and PAHs.

To analyze the microplastics released from CBs, the membranes were observed under Zeiss Primotech microscope (Carl Zeiss Ltd., Cambridge, UK) at magnification levels of 5x, 10x, and 20x. Additionally, scanning electron microscopy(SEM) was used for micro-morphology detection. For PAHs analysis, the 400 mL samples were spiked with 1.0 μL of recovery surrogates, a mixture solution consisting of five deuterated PAH compounds (Naphthalene-D8, Acenaphthene-D10, Phenanthrene-D10, Chrysene-D12, and Perylene-D10) purchased from Dr. Ehrenstorfer, Germany, at a concentration of 200 mg/L. Liquid-liquid extraction was performed using a separation funnel, where 50 mL of Dichloromethane was added three times to extract PAHs. The extract were concentrated and purified and the internal standards (hexamethylbenzene) were added into the final samples before using a gas chromatography-mass spectrometer (GC-MS, Agilent, 7890 NGC-5975MSD) equipped with a fused quartz capillary chromatographic column (DB-5MS) for PAHs detecting (Xing et al., 2016; Chen et al., 2022). To ensure quality, procedural blanks and parallel samples were used in each experiment batch for pretreatment and GC-MS analysis. Solvent blanks and QC standards were used to check for interferences and cross-contaminations, with no significant peaks detected. PAH analysis in parallel samples had <20% relative deviations, within an acceptable error range. Recoveries of deuterated PAH surrogates ranged from 61.8% to 83.2%, and results were corrected accordingly. MDLs of PAHs in samples ranged from 0.02 to 1.80 ng/L. For heavy metal analysis, 10 mL sample was acidified by 1 mL of 1 M HNO_3 and analyzed using Perkin Elmer ICP-MS NexIon 350D. Procedural and reagent blanks were run to check for interference. Multielement calibration standards were prepared from PerkinElmer Pure standards diluted in 5% HNO_3. Calibration range was 0.1 μg/L to 100 μg/L, with a regression statistic requirement of >0.9990. Quality control samples met accuracy and precision criteria (<5%). Instrument parameters were optimized for analysis (Hineman et al., 2010).

3 Results and discussion

3.1 CBs densities

A total of 10,591 CBs were found across all sampled sites, consisting of 3156 on the beach route (1,223 CBs and 0.059 ± 0.002 CBs/m^2 in workdays, 1,933 CBs and 0.094 ± 0.003 CBs/m^2 in weekends), 2,971 on the park route (1,095 CBs and 0.091 ± 0.006 CBs/m^2 in workdays, 1,876 CBs and 0.156 ± 0.013 CBs/m^2 in weekends) and 4,464 on the market route (2,014 CBs and 0.090 ± 0.010CBs/m^2 in workdays, 2,450 CBs and 0.108 ± 0.005 CBs/m^2 in weekends) (Figure 1). The higher number of CBs littering on weekends can be attributed to increased outdoor activities during days off. Thus, it suggests the existence of a positive feedback loop, where increased outdoor activities lead to more littering, including an increase in the number of CBs (Tesfaldet et al., 2022).

The highest average density of CBs litter was observed in the park area, with a value of 0.124 items/m^2. In comparison, the beach area had a density of 0.077 items/m^2, and the market area had a density of 0.099 items/m^2. Previous research has indicated that beaches tend to accumulate higher concentrations of litter compared to urban areas (Geyer et al., 2017), primarily due to natural factors such as wind flow and management factors such as the lack of waste management (Esquinas et al., 2020; Okuku et al., 2022; Xiong et al., 2022). However, the results of this study showed a higher density of CBs in urban areas compared to the beach area. This finding is consistent with the findings observed in Santos city, Brazil, where urban areas exhibited a higher CBs density of 0.25 items/m^2 compared to beaches with 0.20 items/m^2 (Lima et al., 2021). A similar pattern was also observed in Mazandaran city, Iran (Yousefi Nasab et al., 2022).

To investigate whether there were significant differences in the densities of littered CBs between workdays and weekends in different areas, unpaired t-test analysis was performed using GraphPad Prism 9 (GraphPad Software, San Diego, CA, USA; http://www.graphpad.com). The results revealed a statistically significant difference ($p = 0.0049$ in the beach, $p = 0.0238$ in the park) between workdays and weekends in the beach and park areas, while no significant difference ($p = 0.1265$) was found in the market area at the 0.05 significance level. These findings may suggest that during our investigation period, the beach and park areas have higher footfall on weekends than on workdays, and the market always has a higher footfall as the agricultural market is an essential place for people's daily lives.

3.2 CBs brands

A total of 35 cigarettes brands were found among the 10,591 CBs collected in these three survey routes by searching for printed logos, designs or words. The most founds brands were HongTaShan (11.4% on the beach route, 10.7% on the market route and 8.9% on the park route), NanJing (8.7% on the beach route, 9.5% on the market route and 11.8% on the park route), ChangBaiShan(8.3% on the beach route, 9.3% on the market route and 8.5% on the park route) and GoldenLeaf (7.1% on the beach route, 5.9% on the market route and 10.3% on the park route). The top 20 most abundant CBs brands were illustrated in Supplementary Figure 3. On the beach route, 15.3% of CBs were in a late-stage of degradation or too dirty to identify the brand, while on the market route and park route, these figures were 12.7% and 10.5%, respectively. It is noteworthy that the percentage of unidentifiable CB brands in coastal environments was lower than that reported by Lima et al.

(2021) in Santos beaches (53.9%), and the results in urban environments were lower than those reported by Ribeiro et al. (2022) in Niterói (27%). These findings suggest that there are regional differences in the degradation state of CBs, which may be attributed to factors such as solar intensity, winds, currents, and rivers that can affect the breakdown of CBs.

3.3 The presence of heavy metals, PAHs, and microplastics in CBs leachates

HongTaShan (H) exhibited the highest abundance, while ShuangXi (S) had the lowest abundance among the total collected CBs. Based on our investigation, the price of H was 10 RMB, while S was priced at 15 RMB. Considering the widespread presence of H CBs in the environment of Dalian city, we aimed to explore the environmental risks associated with these CBs by examining the contents of PAHs, heavy metals, and microplastics in their leachates. We also compared the CBs from S as a reference.

The heavy metal concentrations of CBs from the two brands are shown in Figure 2. A total of 17 types of heavy metals were detected in H CBs, while S CBs contained 15 types (excluding La and Dy). The results indicate variations in heavy metal concentrations in the CBs of both brands. For example, the observed concentrations of Mn were 297 μg/L (S CBs) and 395 μg/L (H CBs), whereas Pb concentrations were 2.17 μg/L (S) and 0.34 μg/L (H). Generally, Mn and Dy exhibited the highest and lowest average concentrations, with values of 346 μg/L and 0.01 μg/L, respectively, across the CBs of both cigarette brands. Overall, the average concentrations of different metals in the studied cigarette brands followed the order of Mn > Zn > Al > Ba > Rb > Cu > Cd > Ni > Ti > Cr > Pb > Zr > Co > Hg > U > La > Dy. Among the two brands investigated, CBs from the S brand had the lowest total metal concentration at 617.1 μg/L, while CBs from the H brand had the highest total metal concentration at 642.4 μg/L. Considering the price difference between the two cigarette brands, this result suggests that CBs from higher-priced cigarettes tend to have lower total metal concentrations. However, further research is needed to determine the correlation between total metal concentration and cigarette price.

In previous studies, it was confirmed that there are differences in heavy metal concentrations in CBs among different brands (Dobaradaran et al., 2017; Akhbarizadeh et al., 2021a; Soleimani et al., 2022). The mean concentration levels of metals in CBs ranged from 0.005 to 142 μg/cig (Soleimani et al., 2023b) and our results fall within this range. Metals can be introduced into cigarettes through various substances used during the cultivation and processing of tobacco leaves, such as pesticides, herbicides, and moisture-maintaining agents (Lawal and Ologundudu, 2013). Additionally, the presence of metals in cigarettes can be attributed to the utilization of brighteners on the packaging paper (Soleimani et al., 2023b).

A total of 9 PAHs were detected in both of these two different CBs brands (Table 1). According to our chemical analysis results, the concentration of ∑PAHs in the leachates of H CBs was higher than that of S CBs. There are variations in the concentrations of different types of PAHs between the two CBs brands. For instance, the concentration of Naphthalene exceeded 88 ng/L in both CBs brands, while Anthracene was below 0.5 ng/L. Significant differences ($p<0.05$) were observed in the concentrations of all other detected PAHs, except for Naphthalene. Benz(a)anthracene, Chrysene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a) pyrene, Indeno(1,2,3-cd)pyrene, and Dibenz(ah)anthracene have been categorized as probable human carcinogens (Epa, 2008), and inhaling high levels of Naphthalene may seriously affect our health (Diggs et al., 2011). Studies have shown that approximately 5.0 tons of ∑PAHs can be released from CBs into the environment annually worldwide, causing significant harm to life-forms in ecosystems (Dobaradaran et al., 2019; Dobaradaran et al., 2020). These results indicate that the prevalent CBs types in the Dalian environment may potentially leak more PAHs.

Except leaching toxic metals and PAHs, CBs are composed of cellulose acetate, a type of plastic (Pauly et al., 2002; Novotny et al., 2009). When discarded into the environment, the breakdown of these CBs under the influence of wind, heat, ocean currents, and ultraviolet radiation releases microplastics and nanoplastics (Chevalier et al., 2018; Belzagui et al., 2021). The release of microplastics from all kinds of plastics materials has gained significant attention due to its severe harm to ecosystems such as oceans, land, and rivers, as well as human health (Chevalier et al., 2018; Araujo and Costa, 2019; Esquinas et al., 2020; Yang Q. et al., 2023). For instance, the presence of microplastics and nanoplastics in the intestines and other tissues of animals and humans has been reported, which can lead to altered organismal defense mechanisms and reproductive system, as well as tissue inflammation and rejection (Sharifinia et al., 2020; Lim, 2021). Studies have demonstrated that micro- or nanoplastics can negatively impact the growth and reproduction of zooplankton, as well as reduce the chlorophyll absorption of phytoplankton (Slaughter et al., 2011; Dedman et al., 2022). This disruption of the food chain poses a threat to the balance of the ocean ecosystem.

The microplastics leached into the water environment from these two CB brands were observed in the PC membrane through microscopic examination (Figure 3). Electron microscopy results revealed the presence of nanoscale particles in addition to the plastic fibers. Studies have shown that heavy metals such as Mn, Cr, and Pb

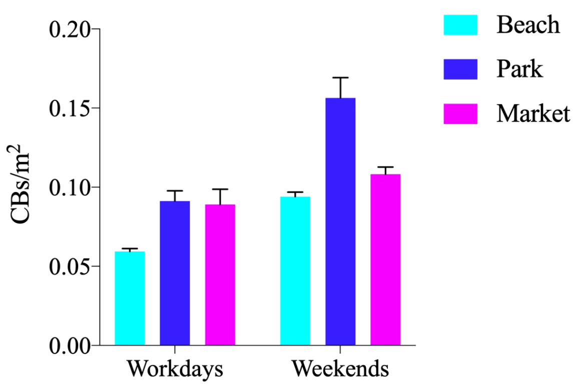

FIGURE 1
CBs density across all sampled sites during the survey period.

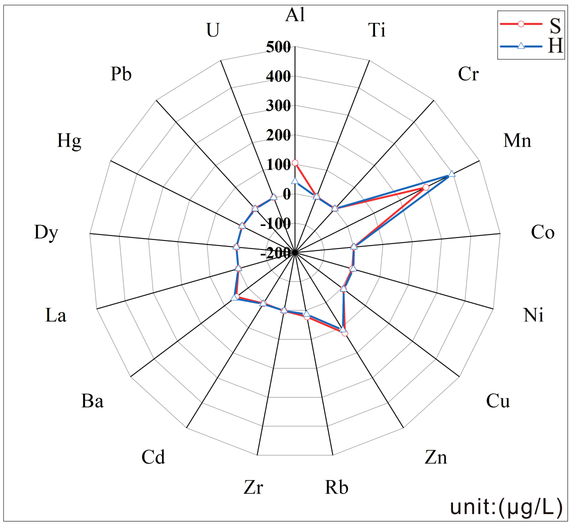

FIGURE 2
The heavy metal concentrations of CBs from the two brands.

TABLE 1 Concentration levels of PAHs (ng/L) in S and H brand CBs leachate.

PAHs	S	H	p-value (between groups)
Naphthalence	92.12 ± 2.02	88.02 ± 0.12	0.0715
Acenaphthylene	11.45 ± 0.52	14.53 ± 0.41	0.0013
Acenaphthene	14.57 ± 0.59	25.67 ± 0.37	<0.0001
Fluorene	12.51 ± 0.51	16.13 ± 0.15	0.0003
Phenanthrene	11.05 ± 0.95	19.76 ± 0.26	0.0001
Anthracene	0.40 ± 0.01	0.46 ± 0.01	0.0011
Fluoranthene	3.00 ± 0.12	6.4 ± 0.05	<0.0001
Pyrene	2.97 ± 0.09	5.87 ± 0.01	<0.0001
Benz(a)anthracene	3.68 ± 0.1	4.16 ± 0.04	0.0013
Chrysene	ND	ND	–
Benzo(b)fluoranthene	ND	ND	–
Benzo(k)fluoranthene	ND	ND	–
Benzo(a)pyrene	ND	ND	–
Indeno(1,2,3-d)pyrene	ND	ND	–
Dibenzo(a,h)anthracene	ND	ND	–
Benzo(ghi)perylene	ND	ND	–
ΣPAHs	157.74 ± 9.19	180.10 ± 8.56	<0.0001

ND, Not Detected.

readily attach to microplastics and nanoscale particles (Chevalier et al., 2018; Dobaradaran et al., 2018a; Akhbarizadeh et al., 2021b). Considering that microplastics and nanoscale particles can disperse into different environments, our findings indicate that the release of CBs-associated microplastics and nanoscale particles may facilitate the spread of toxic pollutants.

China has over 300 million smokers, which accounts for one-third of the world's smoking population, and they consume about 40% of global cigarette production (Lee, 2019). Given that over 6 trillion cigarettes are produced and consumed annually by one billion smokers worldwide, and 75% of these smokers indiscriminately discard CBs (Novotny and Slaughter, 2014; Rahman et al., 2020; Beutel et al., 2021), we can infer that approximately 1.8 trillion CBs enter the environment in China alone each year based on the data. Considering that an average CB contains approximately 0.2 grams of plastic (Green et al., 2014), the annual plastic waste generated by CBs in the environment amounts to 360,000 tons in China. Therefore, these findings highlight the lack of awareness regarding proper CBs disposal in China and the alarming amount of plastic waste and toxic chemical pollutants generated annually by CBs.

3.4 Best practices

The findings of the study, for the first time, suggest that CBs are widely present in Dalian city, with higher densities observed during weekends. This indicates that the lack of strict guidelines for managing CBs and insufficient public awareness in properly handling them, along with human traffic, may be the primary factors contributing to the presence of CBs in the environment. Laboratory experiments conducted to detect CBs pollutants leakage demonstrated the release of heavy metals, PAHs, and microplastics from CBs. Considering the lack of awareness and proper management of CBs, as well as the toxicity and persistence of their leached substances in the environment, discarded CBs will inevitably pose a serious threat to human health, biodiversity, and ecosystem stability.

As a result, urgent measures are needed to avoid plastic pollution caused by CBs. To mitigate the harm caused by CBs, measures should be taken in the areas of education, management, legislation and research. Extensive education campaigns should be conducted to educate people about the proper handling of CBs, and strict guidelines should be put in place, such as imposing penalties on those who litter CBs. Legislation should be enacted to regulate the disposal of CBs. Systematic research is necessary to evaluate the extent of plastic pollution caused by CBs and their impact on human and environments. Scientists must innovate safety equipment and technology in order to minimize the damage caused by CBs. Additionally, governments should incentivize companies to develop non-toxic, biodegradable, and easily recyclable CBs through policies such as subsidies, technological innovation, and resource allocation.

4 Conclusions

The study findings provide the first-ever evidence that the widespread littering of CBs in Dalian, China is a significant problem. A total of 10,591 CBs items were collected during the

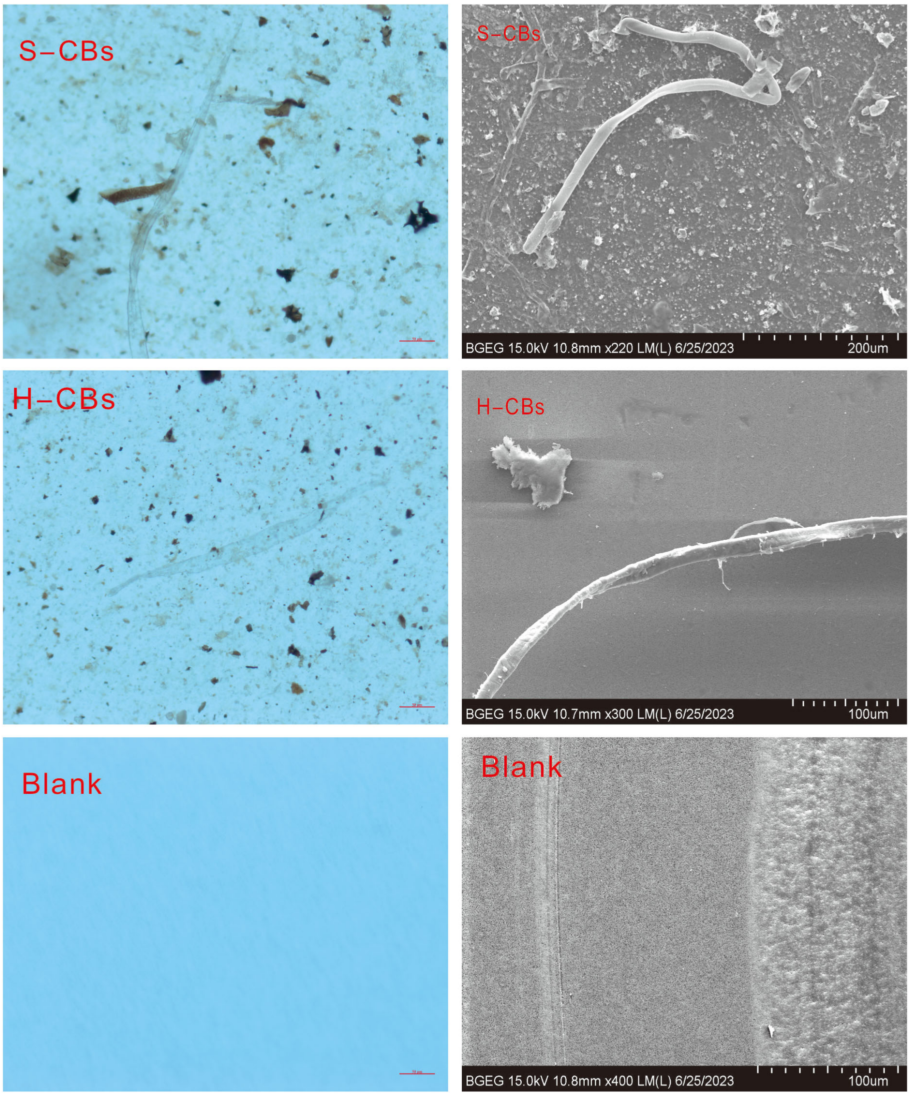

FIGURE 3
Light microscope images of the membrane filters post filtration of CBs from S and H brand taken, as well as the blank membrane at 20 x magnification, along with SEM images of fibers and particles collected from CBs and blank membrane filters.

survey. The highest average density of CBs litter was observed in the park area (0.124 items/m^2), followed by the market area (0.099 items/m^2) and the beach area (0.077 items/m^2). Among these CBs, 35 cigarette brands were identified, and 17 types of heavy metals leached in the highest abundance brand of CBs, with concentrations ranging from 0.02 μg/L (U and La) to 395 μg/L (Mn). The lowest abundance brand of CBs contained 15 types of heavy metals, with concentrations ranging from 0.08 μg/L (U) to 297 μg/L (Mn). A total of nine PAHs, including compounds such as Naphthalene and Acenaphthylene, were detected in both CBs brands, and significant variations in the presence of different PAHs were found among different brands. Microplastics and particles were detected. Based on cigarette consumption data in China, it is estimated that annually 360,000 tons of plastic waste are generated by CBs in the environment. This survey will help raise awareness among the Chinese population about the issue, and the data will be essential for the development of effective environmental protection strategies, such as establishing rigorous management guidelines and promoting innovation and research in reducing the environmental damage caused by CBs.

Data availability statement

The original contributions presented in the study are included in the article/Supplementary Material. Further inquiries can be directed to the corresponding author.

Author contributions

SY: Conceptualization, Data curation, Formal Analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review &

editing. CG: Data curation, Investigation, Methodology, Writing – review & editing. YJ: Data curation, Formal Analysis, Investigation, Methodology, Writing – review & editing. QY: Conceptualization, Data curation, Formal Analysis, Investigation, Methodology, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing.

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Conflict of interest

Authors SY was employed by the company Shenzhen Yuchi Inspection & Testing Technology Co., Ltd.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Supplementary material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fmars.2023.1277402/full#supplementary-material

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