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Science of the Total Environment xxx (xxxx) xxx

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Release kinetics of microplastics from disposable face masks into the aqueous environment

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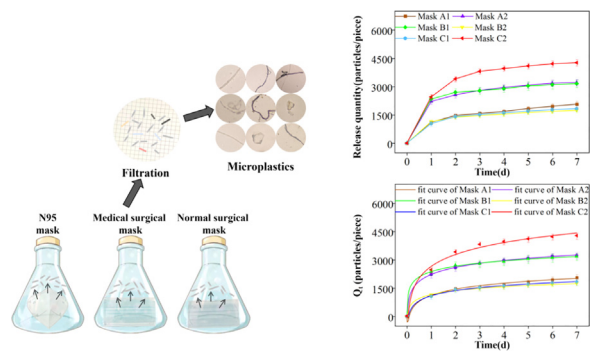
HIGHLIGHTS

- Disposable face masks are identified as one of the sources of microplastics.
- The Elovich equation described the release kinetics of microplastics well.
- The release rate of microplastics was not related with the types of masks.
- Microplastics of <500 μm presented high release quantity and release rate.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 23 August 2021
Received in revised form 19 October 2021
Accepted 9 November 2021
Available online xxxx

Editor: Fang Wang

Keywords:
Microplastics
Release
Face mask
COVID-19

ABSTRACT

Disposable face masks are widely used as primary personal protective equipment to control the spread of the SARS-CoV-2 virus. Disposable face masks have been identified as a source of microplastics and a new threat to the environment when improperly handled. To understand the release of microplastics from discarded masks into water, the release quantities of microplastics from three types of disposable face masks (N95, medical surgical, and normal medical masks) were measured within 24 h and their release kinetics were analyzed over seven days. Results showed that polypropylene microplastics fibers and debris of various colors were released. N95 masks released 801 ± 71–2667 ± 97 microplastic particles/(piece·d), medical surgical masks released 1136 ± 87–2343 ± 168 microplastic particles/(piece·d), and normal medical masks released 1034 ± 119–2547 ± 185 microplastic particles/(piece·d), irrespective of the price, weight, or type of mask. The microplastics were first released fast and then slow. The Elovich equation described the release kinetics ($R^2 > 0.990$), and the release rate did not differ with the type of mask. Microplastics of 100–500 μm and of <100 μm were released in large quantities and at rapid rates. Fiber and transparent microplastics accounted for a large proportion of those released, and their daily release proportion increased with time. Fiber microplastics <500 μm in length were predominant in the microplastics released from disposable face masks, indicating that disposable face masks could be a critical source of these in the aqueous environment. There is an urgent need to take action to implement a waste management system limiting the number of masks entering the environment.

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https://doi.org/10.1016/j.scitotenv.2021.151650
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1. Introduction

Since the first case of COVID-19 was detected at the end of 2019, the COVID-19 pandemic has spread to 212 countries and regions. As of July 2021, >180 million people worldwide have been diagnosed with COVID-19, and the total number of deaths has reached approximately four million (WHO, 2020). Personal protective equipment (PPE) is an accepted mode of self-protection, and is produced on a global scale. During the COVID-19 pandemic, masks are the most important PPE (Chua et al., 2020). The demand for disposable face masks has increased because they are used as a primary personal protective device to control the spread of the virus. In 2020, the monthly global usage of masks reached up to 129 billion (Prata et al., 2020).

The main structure of commonly used disposable face masks is a three-layer nonwoven fabric. The inner layer and outer layers are made of nonwoven polypropylene resin fabrics, and the middle layer is made of meltblown polypropylene fabric with a high melt index. Polypropylene is commonly used in the production of disposable face masks due to its low cost and ease of processing (Chua et al., 2020). Ineffective waste management systems have caused masks to become a new source of microplastics and litter pollution, and a new threat to the environment (Canning et al., 2020; Fadare and Okoffo, 2020; Selvakumar et al., 2021).

Many disposable face masks have been found in rivers and oceans (Saddam et al., 2020; Muhammad et al., 2021). Masks can release microplastics due to photodegradation, weathering, corrosion, and immersion in water (Chamas et al., 2020; Chowdhury et al., 2021), and have been identified as a source of microplastics (Feng et al., 2020; Aragaw, 2020; Anastopoulos and Pashalidis, 2021). Quantifying the release of microplastics from masks is crucial. Researchers have carried out simulation experiments and found that aging due to mechanical action and natural exposure caused the destruction of the mask structure, and lead to the release of microplastics (Morgana et al., 2021; Shen et al., 2021). Chen et al. (2021) found that abrasion and aging during mask use of promoted the release of microplastics, and the middle layer of the mask released more microplastics than did the outer and inner layers (Ma et al., 2021; Wang et al., 2021a, 2021b; Wu et al., 2022). In addition, Sullivan et al. (2021) found that masks not only released microplastics, but also released harmful chemicals such as heavy metals (Pb, Cd and Sb) and organic pollutants. Wearing masks also poses risks of microplastic inhalation and ingestion (Li et al., 2021). Kutralam-Muniasamy et al. (2022) emphasized that although PPE has played an important role in shaping public health, plastic pollution from face masks has become a major environmental and health concern.

The environmental problems caused by COVID-19 can bring attention to waste management systems and create an opportunity to move towards a circular economy (Mekonnen and Aragaw, 2021). Aragaw (2021a) proposed that waste management systems can be designed to reduce pollution from PPE and sustainable management practices can be established to prevent PPE from entering into water bodies. A feasible solid waste management system is also conducive to eliminating mask waste outflow into the environment and reducing pollution (Aragaw, 2021b). It is necessary to investigate the release characteristics of microplastics from masks in order to evaluate pollution risks and improve the management of mask waste. However, few studies are available investigating the kinetics of microplastics released from various brands and types of disposable face masks into the aqueous environment. During the COVID-19 pandemic, mask waste has increased sharply, and it is extremely important to quantify pollution caused by masks.

In this study, we identified the microplastics released from different brands and types of masks, and measured their quantities. We also analyzed microplastic release kinetics and described the microplastics released from masks, including their length, shape, and color, and how these changed over time. This study contributes to a deeper understanding of the mechanisms underlying microplastic release from disposable face masks, and provides a basis for estimating microplastic pollution from mask waste.

2. Materials and methods

2.1. Materials

Twelve brands of three types of disposable face masks were purchased by pharmaceutical corporation, China, and are described in Table 1. Four brands of N95 masks, four brands of medical surgical masks, and four brands of normal medical masks were used in the experiments. There were no respirators on any masks, and all masks met the implementation standards for medical devices in China (Table S1). All masks were new and in an intact and unbroken state, and nose bridges and other accessories were not removed.

2.2. Microplastic release test

A 24-h release experiment was carried out first to compare the release quantity of microplastics among 12 brands of three types of masks. The deionized water used in the release experiment was filtered through a 0.45 μm cellulose ester membrane, and the glassware was rinsed with deionized water before use. A mask sample was placed in a 250 mL conical flask, and 200 mL deionized water was added. The conical flask was placed on a shaker and oscillated at a speed of 220 r/min for 24 h. The mask and the flask were rinsed twice with 400 mL deionized water after shaking. The microplastics released into the water were collected through filtration with a 0.45 μm cellulose ester membrane. The filter membrane was dried in order to evaluate mass loss after filtration. Based on these results the mask with the highest release quantity and the mask with the lowest release quantity of each type were selected, and six brands of three types of masks were used in the release kinetics experiment on the subsequent six days. The quantities of microplastics with different lengths, shapes, and colors were recorded each day. Blank groups without masks were set up to monitor contamination during the experiment. Three replicates were used for each sample in all release experiments.

Three models suitable for describing the releasing process of particles from solid material in liquid (Shadpour and Masoud, 2019; Racovita et al., 2016; Inglezakis et al., 2019), were used to analyze the release kinetics of microplastics (Lü et al., 2007; Wang et al., 2021a, 2021b).

$$\text{Parabolic diffusion equation: } Q_t = a + bt^{1/2} \quad (1)$$

where t represents time (d), Q_t represents the microplastic quantity released at time t (particles/piece), a represents the microplastics released initially (particles/piece), and b represents the rate constant.

$$\text{Power function equation: } Q_t = at^b \quad (2)$$

where t represents time (d), Q_t represents the microplastic quantity released at time t (particles/piece), a and b represents the rate constant.

$$\text{Elovich equation: } Q_t = a + blnt \quad (3)$$

where t represents time (d), Q_t represents the microplastic quantity released at time t (particles/piece), a and b represents the rate constant.

2.3. Microplastic analysis

The microplastics trapped on the filter membrane were observed using an optical microscope (Olympus CX23), and the images were captured using the microscope's digital camera module. Microplastic counts were measured in accordance with De et al. (2018), and S-Eye software was used to record their length, shape, and color. A typical microplastic released from the mask was clamped on a glass slide

Table 1
Description of 12 brands of disposable face masks.

Brand No.	Type	Standard	Weight (mg)	Price (CNY)	Layer	Color	Other description	Manufacturer
Mask A1	N95 mask	GB19083-2010	5.43	1.56	Four layers	White	Sterile, the products are composed of PP spunbond non-woven fabric, melt-blown non-woven fabric, and hot-air cotton, filtration rate ≥ 99%	China, Jiangsu Xianyao Medical Equipment Co., Ltd.
Mask A2	N95 mask	GB19083-2010	4.21	1.84	Four layers	White	Sterile, inner and outer layers of PP non-woven fabric, middle layer of PP meltblown fabric, filtration rate ≥ 95%	China, Henan Yubei Eisai Co., Ltd.
Mask A3	N95 mask	GB19083-2010	4.97	2.15	Four layers	White	Sterile, the product is made of non-woven fabric (PP) by cutting and sewing, filtration rate ≥ 99%	China, Nanchang Chaoyang Medical and Health Products Co., Ltd.
Mask A4	N95 mask	GB19083-2010	4.26	1.75	Four layers	White	Sterile, the mask body is made of non-woven fabric (PP) and filter material, filtration rate ≥ 95%	China, Henan Yiren Medical Equipment Co., Ltd.
Mask B1	Medical surgical mask	YY/0469-2011	2.95	0.44	Three layers	Blue	Sterile, inner and outer layers are PP non-woven fabric, the middle layer is PP melt-blown fabric, filtration rate ≥ 95%	China, Jiangsu Huicheng Medical Technology Co., Ltd.
Mask B2	Medical surgical mask	YY/0469-2011	3.27	0.60	Three layers	Blue	Sterile, inner and outer layers are PP non-woven fabric, the middle layer is PP melt-blown fabric, filtration rate ≥ 95%	China, Qingdao Hainuo Biological Engineering Co., Ltd.
Mask B3	Medical surgical mask	YY/0469-2011	3.62	0.46	Three layers	Blue	Sterile, non-woven fabric 66% (PP), melt blown fabric 34% (PP), sterile, three-layer structure, filtration rate ≥ 95%	China, Hunan Jiuhong Medical Technology Co., Ltd.
Mask B4	Medical surgical mask	YY/0469-2011	2.73	0.09	Three layers	Blue	Sterile, inner and outer layers are PP non-woven fabric, the middle layer is PP melt-blown fabric, filtration rate ≥ 95%	China, Nanchang Demeikang Medical Equipment Co., Ltd.
Mask C1	Normal medical mask	YY/T0969-2013	3.17	0.99	Three layers	Blue	Non-sterile, inner and outer layers of PP non-woven fabric, middle layer of PP meltblown fabric, filtration rate ≥ 95%	China, Henan Chaoya Medical Equipment Co., Ltd.
Mask C2	Normal medical mask	YY/T0969-2013	3.31	0.28	Three layers	Blue	Non-sterile, inner and outer layers of PP non-woven fabric, middle layer of PP meltblown fabric, filtration rate ≥ 95%	China, Jining Aide Biotechnology Co., Ltd.
Mask C3	Normal medical mask	YY/T0969-2013	3.52	0.58	Three layers	Blue	Non-sterile, inner and outer layers of PP non-woven fabric, middle layer of PP meltblown fabric, filtration rate ≥ 95%	China, Xiantao Dingcheng Nonwoven Products Co., Ltd.
Mask C4	Normal medical mask	YY/T0969-2013	2.87	0.09	Three layers	Blue	Non-sterile, mask filter structure made of PP, filtration rate ≥ 95%	China, Guangdong Guangsu Technology Co., Ltd.

with tweezers. Its polymer composition was identified using a Raman microscope (LabRAM HR800), and its Raman spectra were recorded from 100 cm^{-1} to 4000 cm^{-1} with a laser wavelength of 532 nm.

2.4. Quality assurance and quality control

To avoid contamination, particle-free nitrile gloves and laboratory coats were worn during the experiment. The water used in the experiment was filtered through a 0.45 μm filter, and the glassware used in the experiment was rinsed with deionized water before use. The filtration process was carried out in an ultra-clean environment, and the top of the filtration device was sealed with aluminum foil. Blank controls were tested using the same process as the experiment groups, and the quantities of microplastics detected in the blank controls were subtracted from the results of the experimental groups.

2.5. Data analysis

Statistical analysis was performed using SPSS version 26.0 (SPSS Inc., Chicago, IL, USA). Pearson correlation analysis was used to analyze the relationship between the release quantity of microplastics and the price, weight, and mass loss of the masks. The differences in the release of microplastics among the brands and types of masks were analyzed using one-way ANOVAs, and statistically significant difference was set at $p < 0.05$.

3. Results and discussions

3.1. Identification of microplastics

All of the masks sampled in the experiment released fibers and debris observable through an optical microscope (Fig. 1), and indicating that the microplastics were released into the water from the masks. The fibers and debris released by the masks were of various colors (transparent, blue, black, red, brown, and yellow), which was consistent with the findings of Chen et al. (2021) and Sullivan et al. (2021). The characteristic peaks of fibers and debris from the masks were detected at 1334, 1587, and 2948 cm^{-1} (Fig. S1), which matched well with those of polypropylene in the Raman spectrum (Dąbrowska, 2021; Prata et al., 2020). No peak was found at 841 cm^{-1}, perhaps due to biofilm on the surface of the fibers and debris (Sutapa et al., 2018). The fibers and debris released from the masks in the water were made of polypropylene microplastics, the raw materials of masks.

3.2. Quantities of microplastics

The release quantity of microplastics and the mass loss of the 12 brands of masks within 24 h are shown in Fig. 2. Microplastics released from N95 masks, medical surgical masks, and normal medical masks ranged from 801 ± 71–2667 ± 97, 1136 ± 87–2343 ± 168, and 1034 ± 119–2547 ± 185 particles/(piece·d), respectively. Previous studies reported that the release quantities of microplastics from new, unworn masks ranged from hundreds to thousands of particles/(piece·d) (Chen et al., 2021; Morgana et al., 2021), which ranges similar to those in this study. Wang et al. (2021a, 2021b) reported that the release quantity of microplastics from the weathered masks was approximately three times that of new masks. This may be because mechanical action and natural exposure causes changes in mask structure and decreases in mechanical strength, which in turn lead to increased release of microplastics (Wang et al., 2021a, 2021b). Saliu et al. (2021) reported that the release quantity of microplastics from masks treated with accelerated aging was 300 times higher than that of new masks. The average release quantity reached up to 135,000 particles/(piece·d). The masks examined in this study were new and intact, therefore, the release quantity of microplastics in our results may be lower than in real environmental conditions.

The mass loss of 12 brands of masks ranged from 0.293 ± 0.03–0.831 ± 0.035 mg/(piece·d). Mask A4 released the most microplastics and showed the greatest mass loss (Fig. 2). The percentage mass loss of masks in this study ranged from 0.006% to 0.019%. However, if masks underwent aging treatment and were subjected to high mechanical stress, the mass loss percentage increased to between 0.09% and 0.26% (Saliu et al., 2021), suggesting that aging resulted in increased mass loss.

A significant positive correlation was found between the release quantity of microplastics and the mass loss of masks ($r = 0.940$,

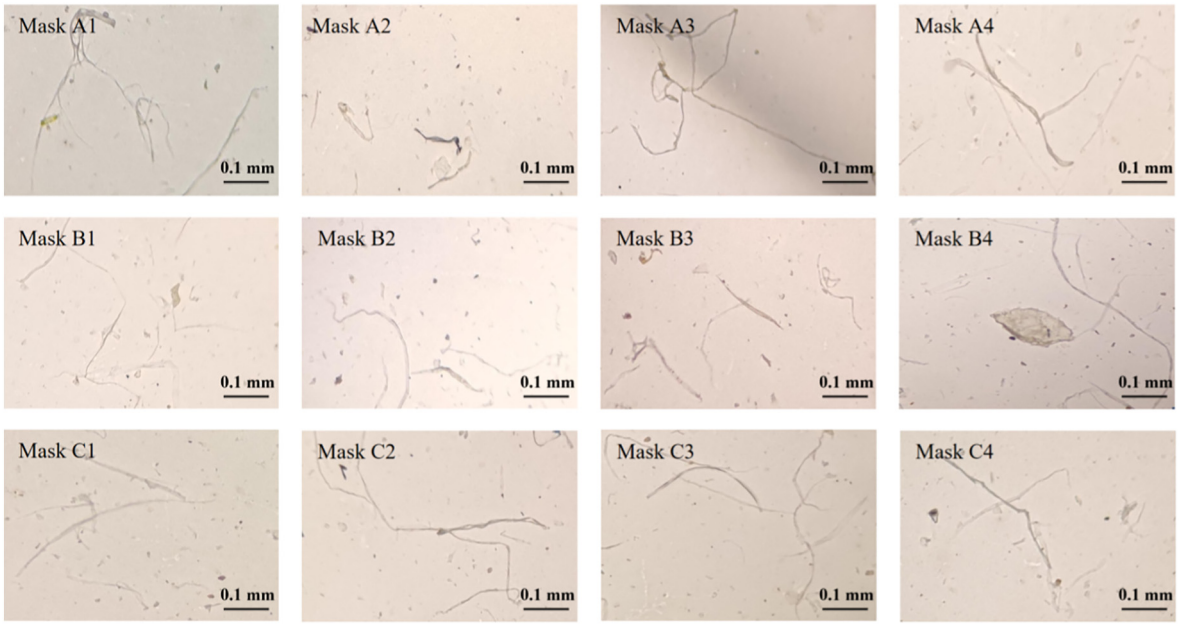

Fig. 1. Optical microscope images of microplastics released from masks.

$p < 0.05$), suggesting that masks can degrade and release microplastics into water. The release of microplastics from masks was the result of breakage and shedding of fibers in nonwoven fabrics (Chen et al., 2021). No significant difference was found between the quantity of microplastics released from N95 masks, medical surgical masks, and normal medical masks ($p < 0.05$), suggesting that the quantity of microplastics released did not vary with the type of mask. N95 masks have a four-layer structure, and the other masks have a three-layer structure (Table 1). Therefore, there was no significant difference in the quantity of microplastics released from four-layered and three-layered masks. Chen et al. (2021) reported that the greatest difference between masks was in the middle layer. Because the masks examined in this study were in the unbroken state and the middle layer was covered by the inner and outer layers, the microplastics we observed may have been released mainly from the inner and outer layers, and this may account for the absence of difference among the different types of masks. In addition, no significant correlation was observed between the quantity of microplastics and the price ($r = 0.017$, $p > 0.05$) or weight ($r = 0.022$, $p > 0.05$) of the masks, suggesting that microplastics release may be not related to the function or quality of masks.

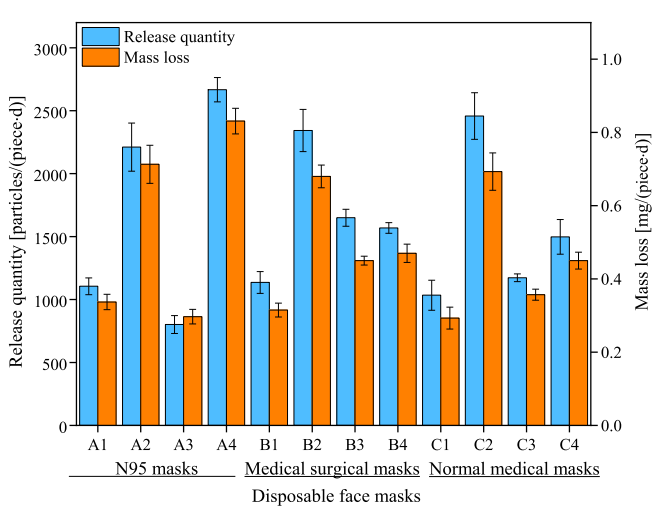

Fig. 2. Release quantity of microplastics and mass loss of masks within 24 h.

3.3. Release kinetics of microplastics

The microplastics from six brands of three types of masks were released according to a similar trend (Fig. 3). The cumulative release quantities of microplastics increased from 1034 ± 119–2457 ± 135 particles/piece on the first day to 1737 ± 82–4270 ± 185 particles/piece on the seventh day (Fig. 3a). Microplastics release was rapid with the increase in release quantity on the first day (Fig. 3b–d). This may be because microplastics on the surfaces of the masks can easily fall off in the water, causing the quantity of microplastics to increase rapidly on the first day. After the first day, the release rate decreased, and the release quantity increased slowly. However, the maximum release quantity was never observed in this study. This may be because the number of easily released microplastics decreased with time, but at the same time the stable microplastics on the masks were released slowly due to aging caused by stir and immersion in water. Wu et al. (2022) found that mechanical abrasion was critical to the release of microplastics from masks, and the quantity of microplastics released into both water and sediment increased rapidly during the first 1–3 days and then gradually slowed. Kutralam-Muniasamy et al. (2022) reported that aging caused by sunlight and mechanical wear broke the masks into millions of microplastics within a few days. Therefore, the gradual aging and decomposition of unbroken masks in the environment may increase the release of microplastics over time, and the release kinetics of microplastics with different aging processes should be of concern in the future.

The release kinetics of microplastics from masks was fitted to the three models, and the model parameters are shown in Table 2. The determination coefficients R^2 of all samples in the Elovich equation and of most samples in the power function equation were >0.990, however, most of those in the parabolic diffusion equation were <0.990. The Elovich equation described the release process of microplastics from the masks best, followed by the power function equation. The Elovich equation is an empirical equation and can describe a complex reaction (Luo et al., 2021; Wang et al., 2021a, 2021b). The release process of microplastics can be divided into two stages, the release of microplastics on the surface of the masks (the first stage) and the migration of

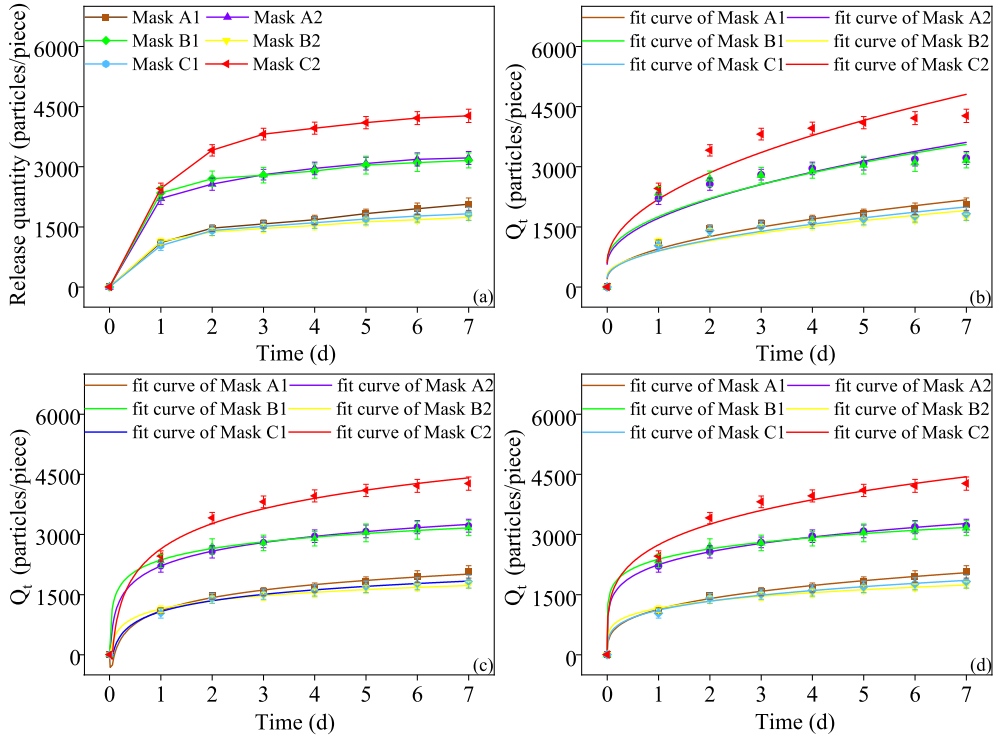

Fig. 3. Quantity of microplastics (a) and fitg curves from the parabolic diffusion equation (b), Elovich equation (c), and the power function equation (d) for microplastics released from masks.

microplastics inside the masks to the surface (the second stage). The good fit of the Elovich equation indicated that the microplastics attached to the mask surface were distributed heterogeneously. The power function equation also described the release process of microplastics (Fig. 3d, Table 2). The values of *b* in the power function equation were all <1, indicating that the release rate of microplastics decreased exponentially with time, as indicated by the derivative of the power function equation (Arzhang et al., 2021). The release rate of microplastics was also proportional to the value of *a*. The greater the release rate, the higher the value of *a*. Therefore, Masks C2, B1, and A2 showed high release rates, whereas Masks A1, B2, and C1 showed low release rates. This also suggested that the release rate of microplastics did not vary with the type of masks. According to the Elovich equation, the cumulative quantity of microplastics released from N95 masks, medical surgical masks, and normal medical masks ranged from 2691 to 4024, 3759–2160 and 2403–5734 particles/piece respectively, after one month.

3.4. Characteristics of microplastics

The length of the microplastics released from masks can be divided into five groups, and their release quantities over time are shown in Fig. 4. The release processes of microplastics from six brands of three types of masks were similar. The quantity of microplastics measuring 100–500 μm was highest on the seventh day (1049 ± 39–2199 ± 139 particles/piece) for all masks, followed by those measuring <100 μm (511 ± 22–1708 ± 138 particles/piece). However, the release quantity of microplastics measuring 500–1000 μm, 1000–2000 μm, and >2000 μm was low. Chen et al. (2021) found that microplastics measuring 100–500 μm were dominant among the microplastics released from masks. Their main structure consisted of interwoven fibers (Ding et al., 2020), and the gaps between the fibers were extremely small, preventing the release of long microplastics. In this study, nanoplastics were not quantified. Morgana et al., (2021) found that the quantities of nanoplastics (0.1–0.5 μm and <0.1 μm) released from masks quantified using flow cytometry was nearly six orders of magnitude higher than the quantities of microplastics found using a microscope. This indicates that nanoscale microplastics accounted for a high proportion of released microplastics and should be considered when estimating the quantity of released microplastics.

The release rates of microplastics measuring <100 μm and 100–500 μm were high on the first day, and subsequently decreased (Fig. 4). The release of microplastics measuring <100 μm nearly stopped after the third day, while microplastics measuring 100–500 μm were released slowly until the seventh day. The release rates of microplastics measuring 1000–2000 μm and >2000 μm were extremely slow, and release nearly stopped on the second day. Hence, the microplastics measuring <100 and 100–500 μm were easily released. Most of the microplastics measuring <100 μm were generated by unevenness in the melt-blown process and the flow of water during the mask manufacturing process (Han and He, 2021; Saliu et al., 2021), and their number was limited. Therefore, the balance was reached after rapid release during the early stage. Microplastics measuring 100–500 μm were released from the masks in the highest quantities, and they were released from the insides of the masks after the rapid release stage. The release rates of microplastics measuring 500–1000 μm, 1000–2000 μm, and >2000 μm were low. This may be because long

Table 2
Parameters and determination coefficients (R^2) from various models used to describe the release of microplastics from masks.

Brand No.	Parabolic diffusion equation			Power function equation			Elovich equation		
	a	b	R^2	a	b	R^2	a	b	R^2
Mask A1	206.1	743.7	0.948	1132.5	0.303	0.997	1096.3	468.9	0.995
Mask A2	566.8	1148.9	0.858	2241.8	0.194	0.999	2210.5	533.1	0.999
Mask B1	659.7	1094.8	0.802	2379.5	0.148	0.999	2361.7	410.7	0.999
Mask B2	282.4	614.1	0.875	1157.4	0.209	0.999	1139.6	300.1	0.999
Mask C1	232.4	667.0	0.916	1110.3	0.263	0.994	1073.6	391.0	0.997
Mask C2	601.5	1588.5	0.892	2739.2	0.248	0.985	2637.2	910.4	0.991

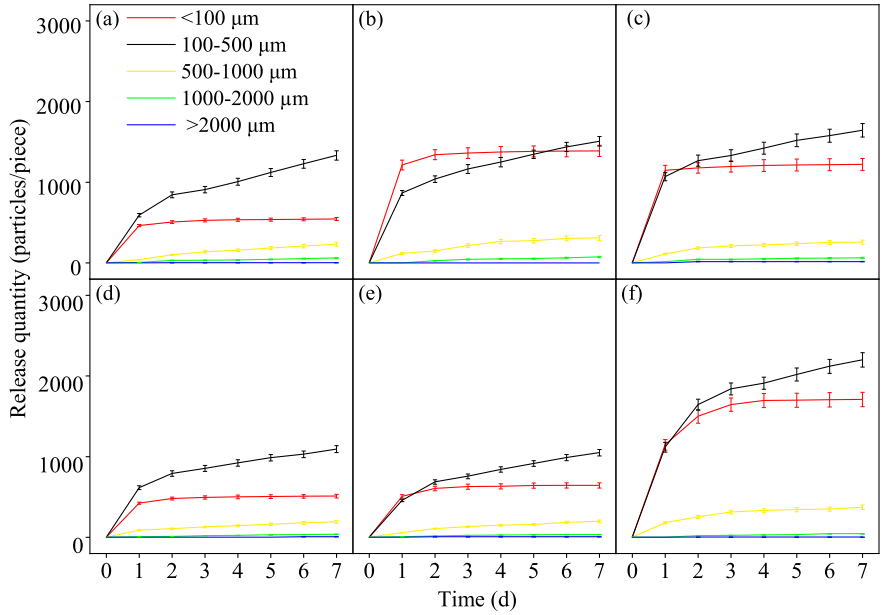

Fig. 4. Release of microplastics of different lengths from masks: (a) Mask A1; (b) Mask A2; (c) Mask B1; (d) Mask B2; (e) Mask C1; (f) Mask C2.

microplastics decomposed into short microplastics under the action of water flow. In addition, the tight fibrous structure of the mask surfaces may hinder the release of long microplastics.

The shape of the microplastics released from the masks over time is shown in Fig. 5. Aggregates were composed of many fibers. These aggregates were classified as fibers and were counted after being separated with tweezers. Fragments and hard-to-separate mixtures (fragments and aggregates) were classified as debris. Fibers accounted for a larger proportion of microplastics than did debris because the masks were interwoven with disordered fibers (Fadare and Okoffo, 2020). The daily release proportion of fibers increased over time, whereas the daily release proportion of debris decreased from 7%–15% to 0% over time, suggesting that the debris were released faster than the fibers. Debris may be a result of impurities mixed into the masks during production and wear, and were mainly distributed on the surface of the masks. Hence, the debris were easily released in the water, and their proportion decreased rapidly.

The microplastics were of several colors and their proportions are shown in Fig. 6. The dominant color was transparent (approximately 80% on the first day), and the proportion the other colors, including black, blue, yellow, brown and red, was low. Since masks are comprised mainly of colorless fibers, it is reasonable that transparent microplastics dominate (Chen et al., 2021). Wu et al. (2022) also found that most microplastics released from different layers of masks were transparent. The microplastics of other colors may have been airborne during production and transportation (Christian et al., 2019). The daily release proportion of transparent microplastics increased with time, whereas the daily release proportion of the other colors decreased from 13%–23% to 13%–3%, indicating that microplastics with colors were released faster than were transparent microplastics. This may be because the proportion of microplastics with colors was small. Most of these microplastics were distributed on the surfaces of the masks and were released quickly due to washing with water.

4. Conclusions

Microplastics were released from different brands and types of disposable face masks in both fibers and debris and in various colors. The release quantity of microplastics from the masks did not depend on the type of masks (N95 masks, medical surgical masks, and normal medical masks). The release quantity of microplastics also did not correlate with the price or weight of the masks, but was positively correlated with mass loss. The release of microplastics from different brands and types of masks occurred rapidly on the first day and slowed gradually. The Elovich equation described the release kinetics of microplastics well. Microplastics measuring 100–500 μm were released in the highest

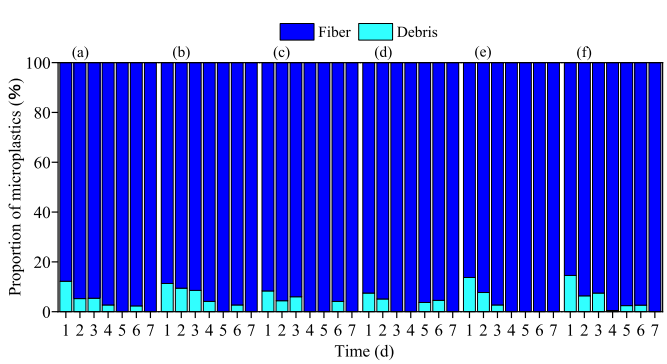

Fig. 5. Proportion of microplastics of different shapes released from masks: (a) Mask A1; (b) Mask A2; (c) Mask B1; (d) Mask B2; (e) Mask C1; (f) Mask C2.

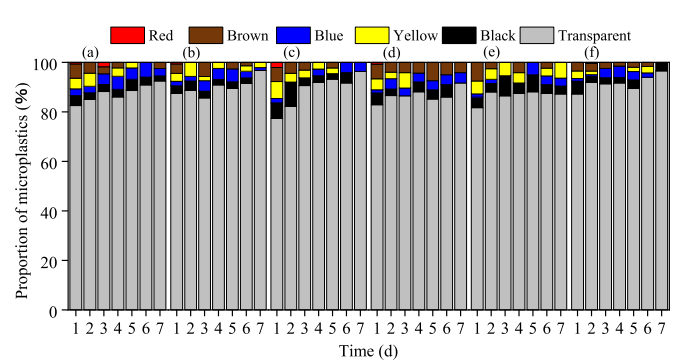

Fig. 6. Color distribution of the microplastics released by different brands of masks each day: (a) Mask A1; (b) Mask A2; (c) Mask B1; (d) Mask B2; (e) Mask C1; (f) Mask C2.

release quantities, followed by those measuring <100 μm, and their release rates were highest on the first day. Microplastic fibers and transparent microplastic accounted for a large proportion of the total microplastics released, and their daily release proportion increased with time. As a country with a large population, China's disposable mask production has reached approximately 200 million per day during the spread of the COVID-19 pandemic. Therefore, there is an urgent need to take action to implement a waste management system that imposes restrictions on the number of masks entering the aqueous environment.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported by the Shandong Provincial Natural Science Foundation (ZR2020MD107).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.151650.

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