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RESEARCH ARTICLE

Different weathering conditions affect the release of microplastics by masks

Ting Zhang[1] · Changrong Zhao[1] · Xi Chen[1] · Angrui Jiang[1] · Zhaoyang You[1] · Kinjal J. Shah[1]

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Abstract

A generation of microplastics caused by improper disposal of disposable masks has become a non-negligible environmental concern. In order to investigate the degradation mechanisms of masks and the release of microplastics under different environmental conditions, the masks are placed in 4 common environments. After 30 days of weathering, the total amount and release kinetics of microplastics released from different layers of the mask were studied. The chemical and mechanical properties of the mask were also discussed. The results showed that the mask released 25141±3543 particles/mask into the soil, which is much more than the sea and river water. The release kinetics of microplastics fit the Elovich model better. All samples correspond to the release rate of microplastics from fast to slow. Experiments show that the middle layer of the mask is released more than the other layers, and the amount of release was highest in the soil. And the tensile capacity of the mask is negatively correlated with its ability to release microplastics in the following order, which are soil > seawater > river > air > new masks. In addition, during the weathering process, the C-C/C-H bond of the mask was broken.

Keywords COVID-19 · Microplastics · Low-load tensile · Compression experiment · Weathering · Disposable masks

Introduction

Since the outbreak of COVID-19 in 2019, personal protective equipment, especially masks, have become a necessity for people around the world to minimize the spread of the coronavirus (Huang et al. 2020). With the spread of COVID-19, the supply and use of disposable masks have gradually increased to unprecedented levels (Zhao et al. 2022). According to the World Health Organizations' previous forecast, the world needs at least 89 million medical masks every month. In addition, 129 billion masks are needed for all populations on earth (Missoni et al. 2021).

Unfortunately, due to poor waste management and a lack of environmental awareness, a large number of post-used masks are thrown away untreated (Cordova et al. 2021; Haque et al. 2021). This raises several issues relating to the disposal and management of discarded masks (Patrício Silva et al. 2021; Prata et al. 2020). Ineffective waste management systems lead to masks as a new source of microplastic pollution and a new threat to the environment (Fadare et al. 2020; Dharmaraj et al. 2021). Disposal of disposable masks has been found to be improper in cities (Akarsu et al. 2021), oceans (Ardusso et al. 2021), rivers (Cordova et al. 2021), beaches (Flores-Cortés and Armstrong-Altrin 2022), and soils (Patrício Silva et al. 2021).

Disposable medical masks are generally made from polymeric materials. Polypropylene is commonly used in the production of disposable masks due to its low cost and ease of processing (Chua et al. 2020). The main structure of the

Ting Zhang and Changrong Zhao contributed equally to this work.

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✉ Zhaoyang You
youzhaoyang@njtech.edu.cn

Ting Zhang
202061224031@njtech.edu.cn

Changrong Zhao
201961124005@njtech.edu.cn

Xi Chen
202161224014@njtech.edu.cn

Angrui Jiang
202161224052@njtech.edu.cn

Kinjal J. Shah
kjshah@njtech.edu.cn

[1] College of Urban Construction, Nanjing Tech University, Nanjing 211800, China

disposable mask is a three-layer non-woven fabric. The inner and outer layers are made of a non-woven polypropylene resin fabric, and the middle layer is formed by ultrasonic welding of filtered melt-blown cloth (Liang et al. 2022). The masks themselves pose a direct biological threat (Gallo Neto et al. 2021). In addition, the release of plastic debris into the environment from masks poses additional environmental problems. Mask waste is subject to a degree of weathering under natural conditions such as wind, water, and UV exposure, which leads to changes in physical and chemical properties and the release of microplastics into the environment (Aragaw 2020). Studies have shown that the type of microplastics released from the masks is polypropylene fibers, which are more biotoxic than conventional granular microplastics (Morgana et al. 2021). The surface of the original polymer becomes rougher with weathering, making the plastic more fragile and more likely to form small chips (Song et al. 2017; Sun et al. 2020). It is worth noting that mechanical abrasion by water and sediments can play an important role in the weathering process (Wu et al. 2022). Chen et al. (2021) reported that the masks usually release more microfibers under water wear and aging. Wang et al. (2021c) reported that the release amount of the middle layer is larger than that of the inner or outer layers. In the context of the new crown epidemic, limitations in management capacity will lead to more mask waste in the environment and release of microplastics (Sangkham 2020). It is estimated that 3.5 million metric tons of masks will be landfilled worldwide in 2020 alone, and these masks will release more than 2.3×10^{21} microplastics (Patrício Silva et al. 2021). Despite the important protective role that disposable medical masks played during the new coronary pneumonia epidemic, microplastic contamination from mask debris has become an unignorable environmental health concern. However, current studies lack data on how masks weathered and released microplastics in different environments. With a surge in mask waste during the COVID-19 pandemic, it is important to quantify the microplastic pollution caused by masks.

To simulate as closely as possible the ability of discarded masks to release microplastics under real-world environmental conditions, we expose commercially available single-use surgical masks to various common outdoor environmental conditions. The difference in the release of microplastics from masks was measured within 1 month after exposure to different environments. By controlling various environmental conditions, the impact of weathering on the release capacity of microplastics was evaluated. We also analyzed the release kinetics of microplastics released from the masks. In addition, by testing the surface morphology and the physical properties of the mask, the reasons for the difference in the release potential of microplastics were investigated. These experiments were conducted to study the release of microplastics in masks under different environmental conditions, and data were collected for supporting the release potential of microplastics under different environmental conditions and providing theoretical support for the environmental burden resulting from the irrational disposal of personal protective equipment during the New Crown pandemic.

Materials and methods

Materials and sample preparation

The disposable medical surgical mask used in the experiment was purchased from a Chinese pharmaceutical company. It consists of an inner layer, a middle layer, and an outer layer. From our previous report, we found that the overall environmental impact of the mask can be determined by calculating the impact of individual layers (Zhao et al. 2022). Therefore, in this study, we have taken into account that all three layers are examined. The seawater required for the experiment was obtained from the Bohai Sea (density 1.02×10^3g/cm^3, pH 8.04), filtered through a 0.22-micron filter membrane and stored in a refrigerator at 4°C. The river water used in the experiment (density 0.99×10^3 g/cm^3, pH 7.28) was collected from the Yangtze River in the Binjiang Wetland Park, Nanjing City, filtered through a 0.22-micron filter membrane and stored in a refrigerator at 4°C. The soil used for the experiment was taken from the surface layer of Nanjing Binjiang Wetland Park, and the soil moisture content was tested after sampling. The soil used for the experiments was pretreated according to the principle of density separation to remove microplastics (Quinn et al. 2017). In brief, the soil was soaked in deionized water for 24 h, and the supernatant was removed. The soaking experiment was repeated twice to ensure that microplastics are removed. Thereafter, the soil was dried at 60°C, and deionized water was added to ensure that the moisture content of the soil retained its initial value.

To simulate the release of microplastics from masks under realistic natural conditions, three layers of an unused mask were placed in simulated sea and river water, air, and soil environments (see Fig. 1). For sea and river water environments, three different layers of mask were dipped into individual 1000-mL beakers and poured to within 1 cm of the beaker mouth (as shown in Fig. 1a), and the beakers were sealed with plastic wrap to prevent the microplastics from escaping into the air. Meanwhile, for the case of the air environments, all three mask layers were placed separately under natural atmospheric conditions (as shown in Fig. 1b). Figure 1c depicts the soil environment where individual layers of the mask were placed in beakers (1000 mL) filled with soil, the depth of the mask in the soil environment being 10 cm below the soil surface. The soil experimental setup

Fig. 1 Photos of the experimental setup for simulating microplastic release in different weathering environments **a** seawater and river water, **b** air, **c** soil, and **d** microplastic fibers adhered to the soil after 30 days of weathering

is placed outdoors to simulate the erosive effects of rain in the natural environment. The weather conditions during the experimental phase are shown in Fig. 2. After 30 days of weathering, the fibers of the weathered mask in the soil are tightly adhered to the soil (as shown in Fig. 1d). Thus, after the 30-day weathering was complete, the masks were removed from all the experimental setups.

In the case of seawater and river water, the microplastics were collected from the beaker and cling film, and the solution was filtered through a 0.22-μm filter membrane, one membrane per 100 mL. The membrane obtained was observed under a light microscope (SOPTOP, EX-30) analyzed at 200× magnification and determine the total amount of microplastics released from this layer of the mask over 30 days. The following procedures were followed for the soil environment study in determining the microplastic release from the mask: (1) remove the 9-cm soil sample below the ground surface (to avoid interference from microplastics in rainwater), (2) collect the soil sample from the upper 1cm of the mask and the lower soil sample, (3) pour 300 mL 30% hydrogen peroxide to digest the soil sample to remove the organic matter in the soil, and (4) add 20 mL 30% hydrogen peroxide to the soil sample. The biochemical reaction was terminated when the soil no longer generated air bubbles. Afterward, all the soil samples were poured into a saturated zinc chloride solution followed by ultrasonic dispersion for 2 h. The supernatant of the solution was vacuumed through a 0.22-μm filter membrane, transferred to a glass petri dish, and dried at 50°C for further analysis. There were two parallel experiments for all experiments.

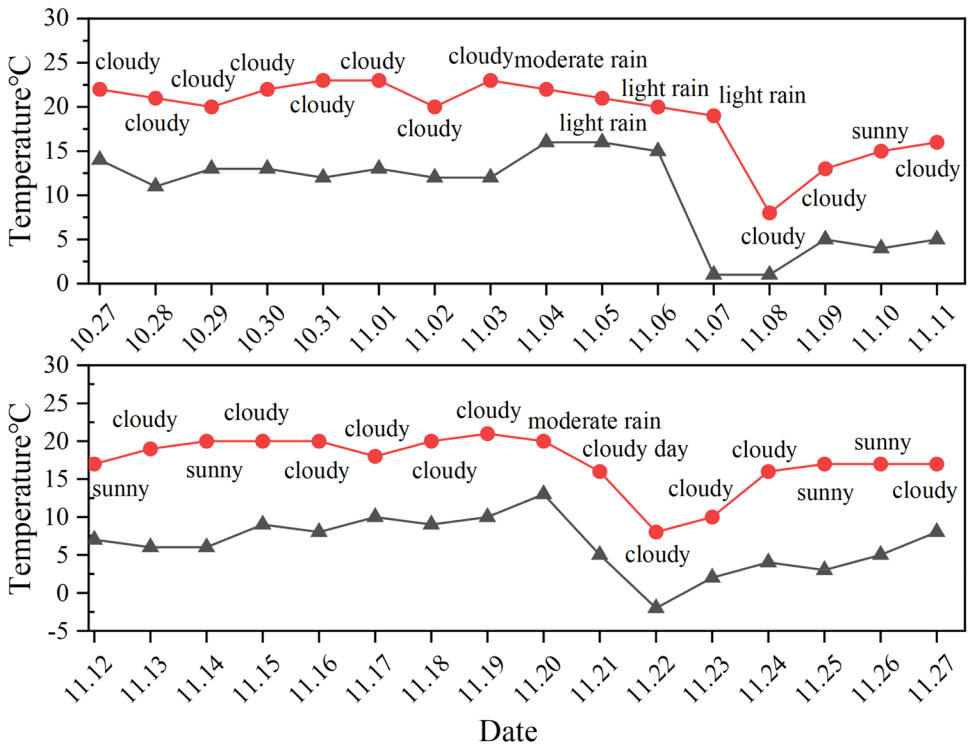

Fig. 2 Weather conditions during the experiment (black line, lowest; red line, maximum daily temperature)

Microplastic release test

To study the potential of different weathering conditions to further release microplastics on masks caused by mechanical wear and tear of beaches and waves in the natural environment, a release experiment was carried out in an environment of water, seawater, and sand. The masks weathered under different conditions were uniformly cut into 3.5×3.5-cm square pieces and placed in a 50-mL conical flask. Twenty-milliliter deionized water or 20 mL deionized water and 6 g sand were placed in the Erlenmeyer flask. After that, the conical flask was placed in a constant temperature shaker and shaken at a constant temperature of 25 °C at 180 rpm for 0–120 h. The following procedures were followed for determining the microplastic release: (1) remove samples from the Erlenmeyer flask the next day, (2) filter the water sample through a 0.22-μm filter membrane, (3) filter the water and sand environment sample into 200 mL saturated zinc chloride solution, (4) sonicate the sample for 2 h to separate the microplastics and sand particles, (5) let the sample stand for another 24 h, (6) pass the solution through a 0.22-μm filter membrane, (7) place the filter membrane in a glass petri dish, and dry it at 50 °C, and (8) place filter membrane under a microscope for microplastic counting and capture images using the microscope's digital camera module.

Three different mathematical models describing the release of solid particles in liquids were used to analyze the kinetics of microplastic release from weathered masks in different environments (Wang et al. 2021a):

Parabolic diffusion model: $Q_t = a + b*t^{1/2}$(1)

Power function model: $Q_t = a*t^b$(2)

Elovich model: $Q_t = a + b*Int$(3)

Among them, t represents time, Qt represents the number of microfibers (particles/single layer) released at time t, and a, b are rate constants.

Quality assurance and quality control

Standard laboratory quality control procedures were followed throughout the experiment. The containers used in the experiment were cleaned with deionized water at least 3 times. To avoid cross-contamination during the experiment, cotton lab coats, cotton masks, and nitrile gloves are worn throughout. The solution used in each step is filtered at 0.22μm before use. Duplicate counts were performed for each sample by different researchers, and deviations were negligible. The microplastics observed in each blank are less than 2%, indicating that the experimental environment is clean enough for ultrafine fiber experiments.

Characterization analysis and mechanical property test of mask

The Fourier transform infrared spectrometer (Spectrum TWO-PerkinElmer, UK) in the attenuated total reflection mode was used to measure each layer of mask material in different weathered environments with the detection wavelength range of 200–600nm. According to the polymer library in the PerkinElmer database (approximately 16,000 reference spectra), PP polymer is used in manufacturing a three-layer disposable surgical mask. A biological optical microscope (EX-30, SOPTOP) was used to magnify the object 200 times to observe the influence of weathering under different conditions on the surface fiber morphology of the mask and the number of microplastics released. The surface morphology of the disposable medical masks before and after weathering was characterized using a scanning electron microscope (SEM, Regulus 8230, Japan).

The mechanical properties of the mask before and after weathering were tested by an intelligent electronic tensile testing machine (Labthink C610M, USA). Every layer of the mask was cut into small pieces with a length of 30mm and a width of 30mm. The mask sample was placed on a low-load tension press and increased the tensile strength at a speed of 5mm/min. The experimental temperature was maintained at 21 °C, and the humidity was maintained at 43%RH. The stop force value and the stop displacement were set at 500N and 950mm, respectively.

Data analysis

Statistical Product and Service Solution 16.0 (SPSS Inc., USA) and one-way analysis of variance (ANOVA) were used to study the comparison of microfiber abundance between each layer, and the statistical significance was set to p-value <0.05.

Results and discussion

Accumulation of microplastics in different weathering environments

To explore the amount of microfiber released by masks in different weathering environments, this study simulated four different weathering conditions: soil, sea, river, and air. As shown in Fig. 2, masks of different layers were placed in the four environments for continuous weathering for 30 days. It can be seen from Fig. 1d that a large number of fibers on the surface of the mask after the weathering of the soil were released to the surrounding environment entangled on the massive soil surface. The microplastic release was also caused by the combined effect of the microbes in the soil due to the hydraulic erosion under rainy weather. As shown in

Fig. 1a, the masks in the water environment all were floating on the liquid surface, and the microfibers on some mask surfaces were released into the external environment as the water was soaked into the mask.

As shown in Fig. 3, the three-layer mask in the soil environment had the most release in one month, reaching 7277±454, 10385±908, and 7479±483 units, respectively. Compared with air and water, the erosion of percolated water in the soil environment and the action of microorganisms in the soil would cause the release of microfibers to increase. The release of microplastics in each layer of the seawater within 1 month was 3314±154, 6331±300, and 3643±378 units, respectively. At the same time, the release of microplastic in each layer of the river water was 3210±132, 5582±280, and 3612±322 units, respectively. Compared with river water, there were more microfibers released in seawater (P value<0.05). This is because the high density of seawater affects the release of microfibers (Liang et al. 2021). By comparing the release amounts of different layers of masks, regardless of the weathering conditions, the release of microfibers by the middle layer was found to be the most, the inner layer is the second, and the outer layer is the least. This is consistent with the findings by Wu et al. (2022). This is very likely because the middle layer of disposable surgical mask is made of velvet fibers of 0.5–10 micron by velvet blasting process, and the surface bonding is not strong (Liang et al. 2021). From the release of microplastics into the soil environment, it can be seen that the microfibers on the soil surface were affected by the external environment and fell off. The materials used for the inner and outer layers are all spun-bonded non-woven fabrics, and the inner layer is designed to be softer due to direct contact with the human skin. Previous studies have indicated that the hydrophobicity of the outer mask is higher to meet liquid repellency requirements (Nongnual et al. 2022). Therefore, the release of microplastics through the inner layer material is relatively high compared to the stronger and more hydrophobic outer mask.

The pictures of the released microplastics can also be seen (Fig. 4). The fibers released from the outer layer and inner layer are thicker, while the fibers released from the middle layer are thinner. Also, the color of the microplastic released from different mask layers is different. The outer layer is blue, the inner layer and middle layer are white fibers, and it is more transparent under microscope observation. This corresponds to the color contrast of different mask layers on the macro. At the same time, there are some holes and fractures on the weathered microplastic surface, which were degraded by the combined action of ultraviolet radiation and microorganisms in the natural environment. Similar findings were observed in our previous findings (Zhao et al. 2021).

Release capacity of microplastics after weathering

Consider the possibility that masks in the soil environment may enter water bodies through groundwater or wind action. Based on research by Liang et al. (2021), we simulated the dynamics of microfibers released into the water environment by masks weathered in different environments under different mechanical wearing and tearing processes. As shown in Fig. 5, the ability of masks to release microplastics in four weathering environments shows a similar trend. That is, as

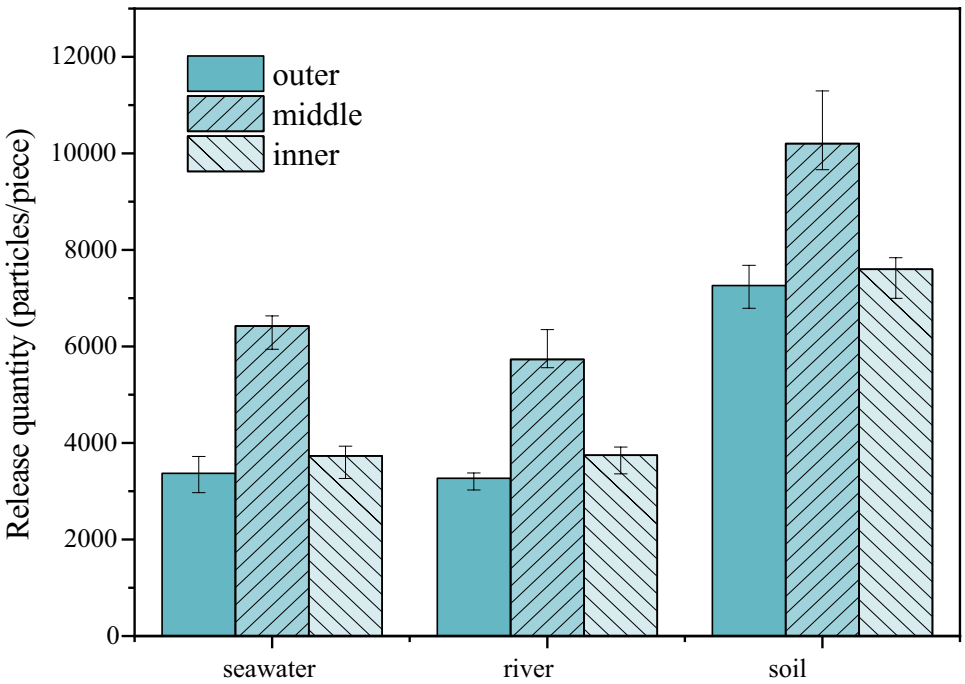

Fig. 3 Total amount of microplastics released in different weathering environments after 30-day time

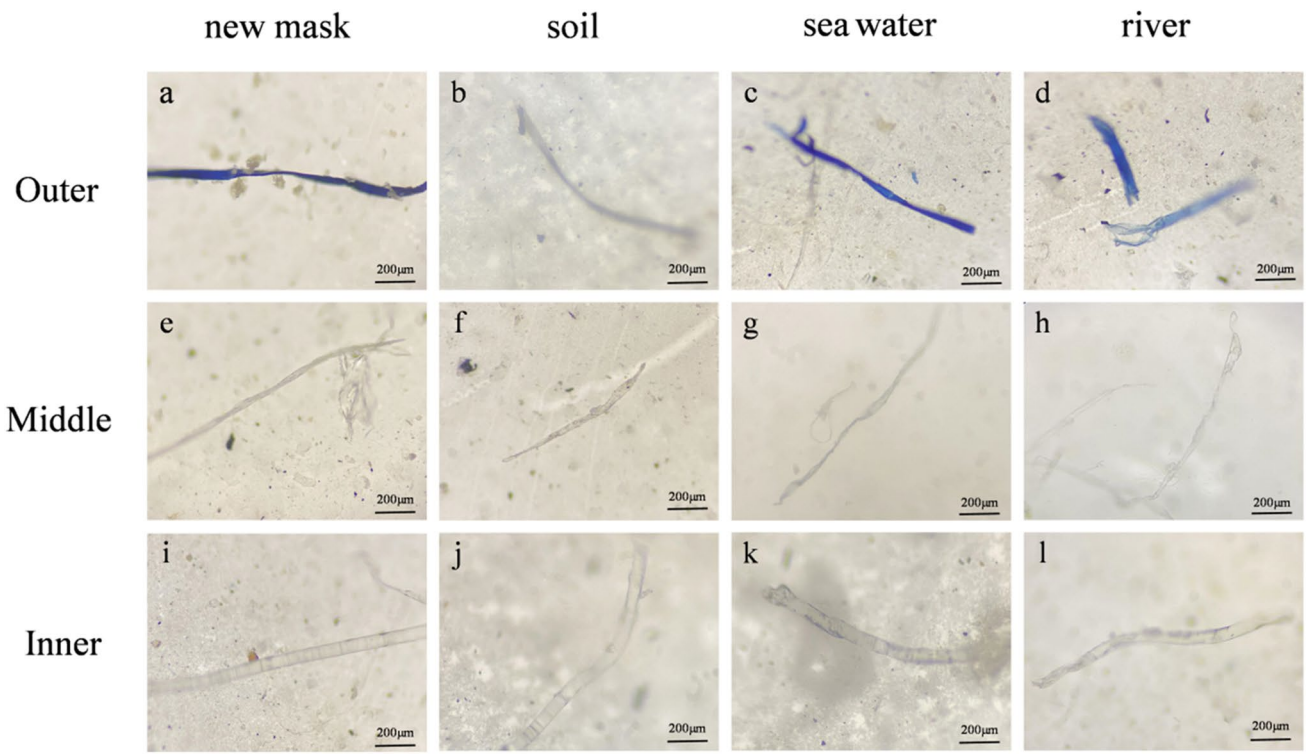

Fig. 4 Different layers of microplastics released after 30-day weathering; **a**, **e**, **i** new mask; **b**, **f**, **j** soil; **c**, **g**, **k** seawater; **d**, **h**, **l** river water

time increases, the rate of release gradually slows down, because after weathering, there are a large number of microplastics that easily fall off on the mask surface. When they are immersed in the water environment, these microplastics fall off rapidly under the action of hydraulic scouring and mechanical wear, resulting in a rapid release of microplastics in the initial stage. Within the release time, the unstable microfibers on the surface of the mask were gradually released, and the release rate slowed down. In addition, the middle layer releases more microplastics than the outer and inner layers, which may be due to the fact that the melt blown process for the middle layer forms thinner and more crushed fibers than the spun bond process used for the inner and outer layers. The amount of microplastics released by aging masks follows the soil> seawater> fresh water> air > unaged masks.

It has been found that after being oxidized by atmospheric oxygen, sunlight, and mechanical wear, masks can break down into millions of microplastics within a few days (Kutralam-Muniasamy et al. 2022). Based on this, the effect of mechanical wear caused by sand on the release of mask microfiber was studied by simulating the common mechanical wear of water and sand. Compared with the samples without sand, the number of microfibrils released by the three-layer mask in sand doubled. It shows that the mechanical wear caused by sand will significantly increase the release of mask microfibers in the environment. We can also see that no matter what hydraulic conditions, the release trend of microfiber in each layer of the mask under different weathering conditions increases rapidly at first and then slows down gradually.

By using three-particle diffusion kinetics models, i.e., Elovich model, power function, and parabolic diffusion, to simulate the amount of microplastic released by the mask, it was found that the correlation result between two variables (number of microfibers and time) via the Elovich equation is the best (see Table R1). The correlation coefficients of most of the samples in this model are greater than 0.97, indicating that under all weathering conditions, the microplastics released by masks at various times were well predicted by the Elovich equation. In addition, it can be seen that the value of the constant a (released particles) of the Elovich equation is increased in all weathering conditions compared to the control, and the constant b, which can be used as an indicator of the transformation rate, shows that the rate of transformation is a very small value in all cases, but higher in river and soil cases compared to the control case, indicating that the transformation rate of microplastics is higher in soil and water weathering conditions than in air and control. Moreover, Elovich equation is an empirical equation used to describe complex reactions. This shows that the release process of microplastics can be divided into two stages. The first stage is the release of microplastics on

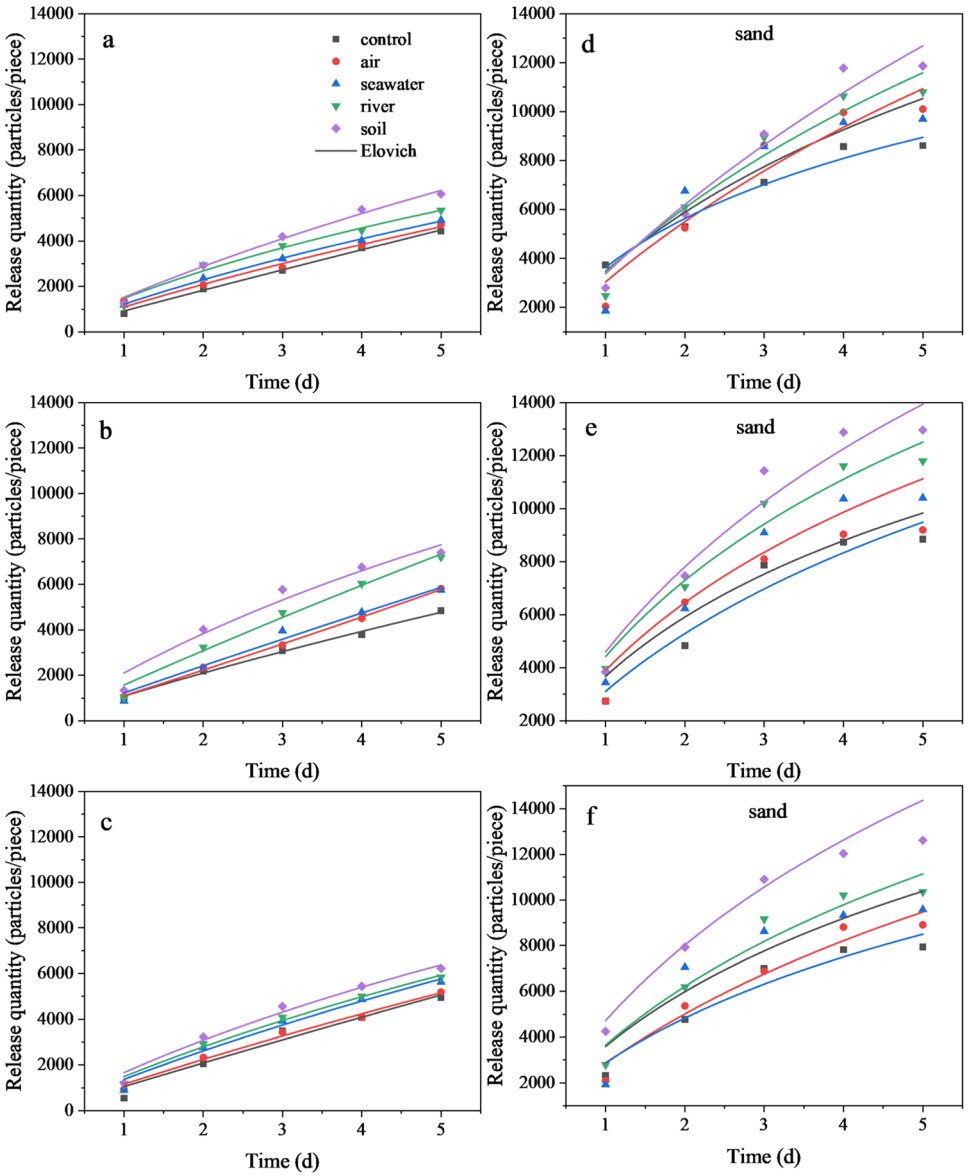

Fig. 5 Fitting curve of release kinetics of mask microplastics; **a**, **d** outer layer; **b**, **e** middle layer; **c**, **f** inner layer. The left column is the experimental group with only water added, and the right column is the experimental group with water and soil added

the mask surface under the action of external force, and the second stage is the migration of microplastics inside the mask to the surface. The Elovich equation fits the data well, suggesting that the microplastics adsorbed on the mask surface were unevenly distributed. According to the release kinetic curve, we can see that weathering enhances the ability of each layer of the mask to release microplastics. The order of enhancement in descending order is soil > seawater > river water > air > new mask (P value<0.05). Comparing the ability of the three-layer structure of the mask to release microplastics, it can be seen that the released amount of the middle layer is 500–2500 particles/single layer higher than the remaining two layers. It further illustrates the role of soil and river environments in promoting the release of microplastics from masks, especially inner masks.

Changes in morphology and mechanical properties caused by weathering

The change of surface texture is the key factor to evaluate the degree of weathering. In this study, the masks before and after weathering were analyzed by a scanning electron microscope (see Fig. 6). From Fig. 6a, f, and k, it can be seen that the texture of the mask changes after its exposure to weather, i.e., air, river, sea water, and soil. The surface of the original mask fiber is smooth (see Fig. 6a, f, k), but there are a few cracks, which are likely caused by the production process (Han and He 2021). However, after a month of natural weathering, similar changes have taken place in the outer and inner layers, that is, the surface of the mask fibers has become rough and has developed more gaps, which is caused by the damage of radiation and other factors to the

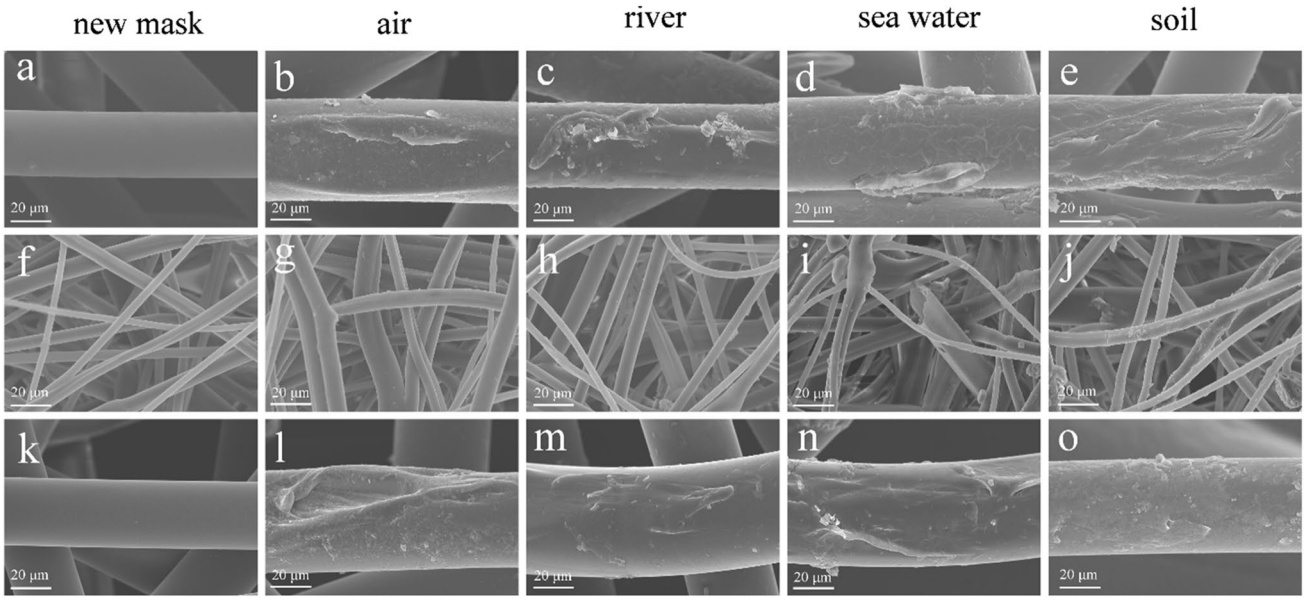

Fig. 6 SEM of the outer, middle, and inner layers of disposable medical masks aged in different environments: **a**–**e** outer layer, **f**–**j** middle layer, and **k**–**o** inner layer

easily bent fiber intersections. The obtain results are in line with reported work by Wang et al. (2021b). At the same time, some particles and fiber fragments were formed at the damaged place. The fiber surfaces of the intermediate layers have a relatively obvious weathering, not only with cracks on the fiber surfaces but also with large clumping. At the same time, compared with the mask fibers weathered in different environments, the fibers on the mask surface weathered in seawater and fresh water are coarser than those weathered in air. These changes may be due to the presence of high levels of microorganisms and chloride ions in the aqueous environment, which promote changes in the surface structure.

In the natural environment, the mask not only releases microfibers but also decomposes into visible sheet plastics with the action of water and wind (Fadare and Okoffo 2020). Through low load tension and compression experiments, we explored the changes in stress-strain curves of each layer of mask after weathering for 30 days under different conditions. As shown in Fig. 7, the new mask can withstand a greater tensile strength, indicating that the material is bonded more tightly. After a month of weathering in the soil, the three-layer material of the mask is less resilient than under other conditions. Compared to new masks, the tensile strength of masks after soil weathering decreases by 50–66%. From top to bottom, the decreasing order of the tensile capacity of masks is new mask > air > river > seawater > soil. This is consistent with the number of microfibers released from the mask. At the same time, it can be seen that the tensile capacity of the middle layer of the mask is much lower than that of the outer and inner layers. When the tensile strength reaches 2.6MPa, all the middle layers break, while the tensile capacity of the outer and inner layers can reach about 6Mpa. This shows that the structure of the middle layer of the mask is relatively loose, which is easily affected and broken in the weathering environment, and microfibers are released from the mask. At the same time, we can find that weathering in various environments not only weakens the tensile capacity but also lengthens the tensile length of the outer layer and middle layer compared with the original mask. This may be due to the attachment of microorganisms and the adsorption of hydrophobic substances and extracellular polymers in the environment on the mask, which makes the mask toughness stronger.

Changes in chemical composition caused by weathering

ATR-FTIR analysis was carried out on three layers of disposable medical surgical masks to analyze the structural changes of mask fibers after weathering. As shown in Fig. 8, there are two peaks in the range of wave number 1300 to 1540 cm^{-1} and four peaks in the range of 2800 to 3000 cm^{-1} for all layers. In this test, the peak at 1456 cm^{-1} is due to $-CH_3$ asymmetric vibration or $-CH_2$ shear vibration, while the peak at 1375 cm^{-1} is the result of $-CH_3$ symmetric deformation. This result also shows that the peaks at 2870 and 2950 cm^{-1} are due to the asymmetric and symmetric tensile vibration of $-CH_3$ (Morent et al. 2008; Ullah et al. 2020). The small peak between 700 and 1200 cm^{-1} is considered to be the characteristic band of polypropylene structure,

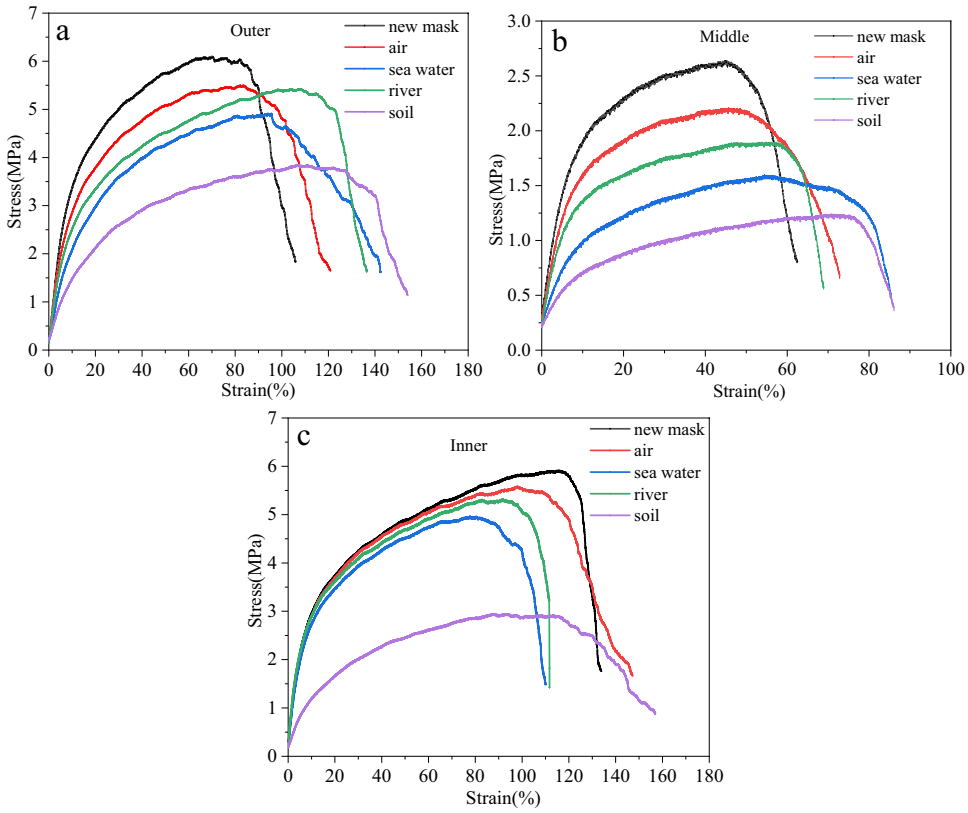

Fig. 7 Low load tension-compression experiments of different layers of masks; **a** outer layer, **b** middle layer, and **c** inner layer

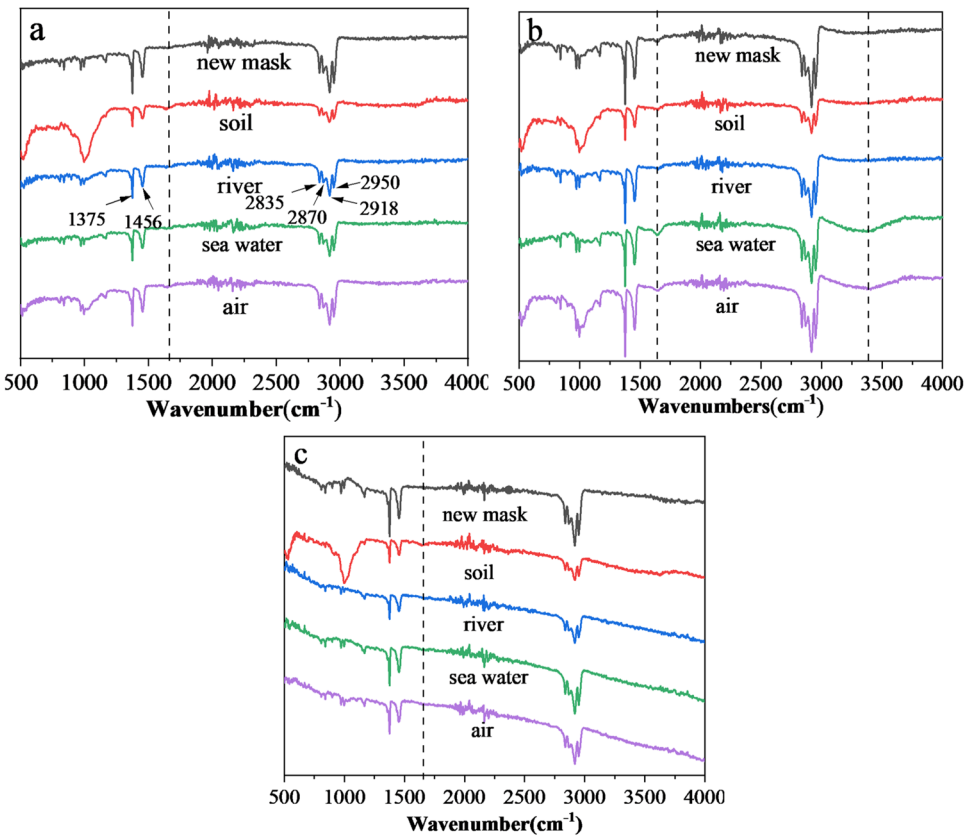

Fig. 8 FTIR of masks under different weathering environments, **a** outer layer, **b** middle layer, **c** inner layer

including asymmetric and symmetric stretching of the C-C bond, rocking vibration of C-H bond, the asymmetric rocking of -CH_3, and asymmetric rocking vibration of -CH_2. Infrared spectrum analysis shows that the three-layer masks are made of polypropylene.

Comparing the spectra of masks weathered in different environments, it was found that a group of small peaks appear at 1600~1750 cm^{-1} at the same time. These small changes show that some hydrocarbon bonds are breaking and forming a double-bond structure (Aslanzadeh and Haghighat Kish 2010; Aslanzadeh and Kish 2005). In addition, it is worth noting that the structure binding ability, slightly weathered by nature, varies with different layers, and the middle layer is most affected, followed by the outer layer and the inner layer. For example, after weathering in seawater, fresh water, and air, the peaks in the outer and middle layers are smaller in the range of 1600–1750cm^{-1}, while the front produced by the samples in the soil is significantly higher. In this experiment, the mask was weathered under natural conditions, which caused the breaks between the C-C bond and C-H bond, generating alkoxy and peroxy groups and further leading to chain fracture or cross-linking. At the same time, some double bond groups, such as carbonyl, were also formed.

Conclusion

In this study, the weathering of disposable masks in four common environments was simulated, and the characteristics of microfiber release were analyzed experimentally. Compared with the new mask, the weathered mask released a significant amount of microfiber, and the order of release ability from largest to smallest is soil > seawater > river > air > new mask. Among the three-layer materials of the mask, regardless of the weathering environment, the middle layer releases the most microfibers, while the outer layer and the inner layer have little difference in release capacity due to their similar structure. After adding sand, the number of microfibers released by the mask doubled within 5 days due to mechanical wear and tear. Three kinetic particle diffusion models were used to simulate the amount of microplastic released by the mask. The Elovich equation was found to be the best model to correlate the amount of microplastics released at different times. The weathering process was used to study the mechanical properties of layered masks under different weathering environments, and it was found that exposure to the environment reduced the tensile strength of masks after soil exposure decreased by 50–60%. In addition, different environmental aging will also change the surface chemical properties of microplastics. Various environmental aging causes the materials to show that the C-C bond and C-H bond break, forming a double bond structure and leading to fiber breaks. The results of this study provide theoretical and data support for the release amount and release potential of microplastics in masks under different weathering conditions.

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Data availability The authors confirm that the data supporting the findings of this study are available within the article.

Declarations

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