

Original Research

Fate of Microplastics Released by Discarded Disposable Masks

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Abstract

The pandemic of COVID-19 has led to a surge increase in the production of masks. Due to the rapid propagation of COVID-19 and the long survival time of plastic surfaces, a large number of masks are discharged into the environment without treatment. In this paper, the release of microplastics (MPs) in nature was simulated by using mask samples irradiated by ultraviolet (UV) light. After 28 days of ultraviolet radiation, part of the main chain of the mask was broken and a large number of transparent MPs fell off. The longer the UV irradiation time, the larger the proportion of small particle MPs. The middle layer of surgical mask is the most difficult to release MPs due to charge treatment, and N95 mask is the most difficult to degrade the inner material.

Keywords: COVID-19, disposable masks, microplastics (MPs), ultraviolet (UV) irradiation

Introduction

COVID-19 virus has been rampant in the world since December 2019, causing a series of severe respiratory syndrome [1, 2]. Scientists around the world have found that COVID-19 virus can be transmitted not only through direct contact, but also through contact with contaminated surfaces/wastes, air/respiratory droplets and feces [3, 4]. Today, the global infection rate is increasing geometrically, and a large number of people have symptoms of infection and died. In this case, the researchers found that masks can control the

spread of the COVID-19 virus, so it is recommended that people wear masks in public places to reduce the risk of virus transmission [5]. The severe epidemic situation makes the mask as a simple anti-virus measure rapidly popularized in many countries. As a disposable consumer goods, the production of masks is growing rapidly in the world. It is estimated that 129 billion masks are needed worldwide every month to stop the spread of COVID-19 [6]. In March 2020, the daily output of masks in China was 10 times that in January 2020, reaching 200 million [7]. Japan's production of masks has also increased rapidly, with orders for masks reaching 600 million in April 2020 [8].

The raw materials of disposable masks are various polymer materials for the production of various plastic products [9]. With the COVID-19 pandemic, all kinds

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of plastic wastes are increasing [10]. India produces 22 kg of plastic waste for every 1000 COVID-19 RT-PCR tests [11]. 1.1 tons of disposable plastic are produced for every 250 tons of medical waste [12]. Due to the characteristics of rapid transmission of COVID-19 and extended survival time on the plastic surfaces, people prefer plastic production rather than recycling [13, 14]. Most of the discarded masks go directly into landfills, fresh water and oceans without any treatment, and were degraded into smaller microplastics (MPs) (plastic debris and particles less than 5mm in diameter) through natural degradation/fragmentation or decomposition [15, 16]. Researchers found that once disposable masks entered the environment, a large number of MPs could be produced in a short time and have the potential to spread across the globe [17]. Recent researchers have found that a large number of disposable masks have been found on the beaches of Hong Kong, the Magdalena River in Colombia and along the highway and drainage of Ile Ife in Nigeria [18, 6, 19]. A large number of masks may enter from beaches to the sea and become available to affect global environment [20]. Therefore, more and more attention has been paid to the large amount of MPs produced by disposable masks entering the environment during the pandemic of COVID-19.

Untreated disposable masks enter the environment and gradually become the main birthplace of MPs after a series of conditions such as wind, light, water and wear [9, 21, 22]. The researchers confirmed this conclusion through the detector of the infrared spectrum [19]. MPs in the environment were difficult to be degraded by microorganisms, so they gradually accumulate in the environment, affecting aquatic organisms, agriculture, forestry and tourism [23]; threatening human health and safety [24, 25]; and posing a serious threat to biodiversity [26, 27]. Therefore, a large number of MPs produced by disposable masks entered the environment during the pandemic of COVID-19. So far, the characteristics and rules of the released MPs from masks in the natural environment have not been systematically studied.

In this study, we used ultraviolet (UV) irradiation to simulate the natural light, and investigated the characteristics of MPs released by N95 masks and surgical mask in water. The release of MPs from disposable masks in natural environment was studied, and the influencing factors of large release of MPs were investigated. To provide a basis for the mandatory recycling and disposal of plastic waste during the epidemic.

Materials and Methods

Preparation of Masks

In this paper, we chose N95 masks (3M9501/9502, USA) and surgical masks (Winner, China) commonly

used during COVID-19 to investigate the released rules and characteristics of MPs in natural environment. Both masks were produced in 2020 to prevent the virus. First of all, the complete new mask was cut into 1 cm × 1 cm, each block was divided into outer layer, middle layer and inner layer. Then the mask was cleaned with non MPs water prepared in the laboratory to remove the MPs particles on the surface of the mask. Finally, the mask was dried under natural conditions and repeatedly used mask during the period of COVID-19 was prepared.

Experimental Design MPs Fall off Caused by UV Radiation

Since light irradiation is the most widely used technology for MPs aging, and it is reported that the polymer of mask can be significantly degraded under UV-A (320-400 nm) and UV-B (280-320 nm) radiation [28]. Put one 1cm × 1cm single-layer mask block into a beaker and add 100 ml non MPs water and a certain amount of glass beads to simulate the friction of mask in nature. Then, the single-layer mask block was irradiated 24 hours a day with UV lamp (1W/m², 365nm) for 28 days. During the experiment, 12 single-layer mask blocks (including 4 outer layers, 4 inner layers and 4 middle layers) of each kind of mask were randomly selected for UV irradiation. Three mask samples (one inner layer, one outer layer and one middle layer) were randomly taken out every 7 days for detection, and the temperature was maintained at 22±3°C. This experiment was repeated three times from March 2021 to May 2021.

Qualitative and Quantitative Analysis of MPs

Take out the sample after UV irradiation, clean it with ultrasonic for 15 minutes (multiple use of masks samples without UV irradiation were used as blank samples), and repeatedly clean the mask sample with 200 ml non MPs water for 3 times. After cleaning, clamp out the sample with clean tweezers and stand for 30 seconds to remove excess liquid from the sample. Collect the cleaning water for three times and filter it with polycarbonate membrane (bore diameter 10 μm, Whatman). The MPs particles falling off by UV light in the cleaning water were trapped on the polycarbonate membrane. After filtration, take the filtered polycarbonate membrane and put it into the oven to dry (40°C) for standby. All the instruments involved in this experiment were cleaned with water without MPs before the experiment to prevent the MPs on the instrument hindering the results.

In order to identify the quantity of visible to the naked eye MPs on the filter membrane (N_f), a stereomicroscope (SMZ1270, Nikon, Japan; 16×) was used to locate and identify the MPs particles by a digital camera. Then the micro-Fourier Transform Infrared Spectrometer (micro-FTIR) equipped with

Attenuated Total Reflection (ATR) (PerkinElmer Spectrum Spotlight 200i, PerkinElmerInc., USA) was used to count, determine the color and measure the size of the MPs particles invisible to the naked eye. Finally, the measurement results were compared with FTIR database to determine the type and structure. In this process, the total number of the MPs particles invisible to the naked eye on the filter membrane (N_2) can be calculated by the following formula:

$$N_2 = N_{20} \times \frac{S_0}{S}$$

where N_{20} is the number of MPs particles of the area detected by micro-FTIR imaging; S_0/S is the ratio of the area observed by micro-FTIR imaging to the area of the filter membrane. The sum of N_1 and N_2 is the total number of MPs plastics in the filter membrane. The specific detection methods refer to Li et al [29].

Results and Discussion

Changes in Structure of the N95 Mask and Surgical Mask under UV Radiation

In this paper, ATR-FTIR was used to analyze the structure changes of each layer of two kinds of masks

after UV irradiation (Fig. 1), and found that the mask material used in the experiment was polypropylene. After 28 days of experiment, each layer had obvious changes, and the changes were similar. Each layer of the two kinds of masks had uniform and obvious absorption bands at 2945 cm^{-1} and 2867 cm^{-1} , and the peak value decreased in varying degrees after UV irradiation, which depended on the stretching vibration of methyl group [30]. After 28 days of UV irradiation, the peak values also decreased at 2920 cm^{-1} and 2836 cm^{-1} , which may be caused by the stretching vibration of methylene group [31]. In addition, the peak values at 1450 cm^{-1} and 1380 cm^{-1} decreased, which proved the efficient oxidation of aliphatic hydrocarbons under UV irradiation. Similarly, C-C stretching, C-H rocking vibration, methyl group asymmetric rocking vibration and methylene group asymmetric rocking vibration also occur in hydrocarbon related components (1164 cm^{-1}) after UV irradiation. It can be seen that UV radiation provides enough energy for C-C and C-H bond breaking to produce alkoxy and peroxy groups. The results showed that the two kinds of masks after 28 days of UV irradiation led to the fracture of part of the main chain and the decrease of molecular weight, caused the mechanical properties of the mask were reduced and a large number of MPs were produced [32]. This is consistent with the previous conclusion that UV can make some groups of polypropylene react and

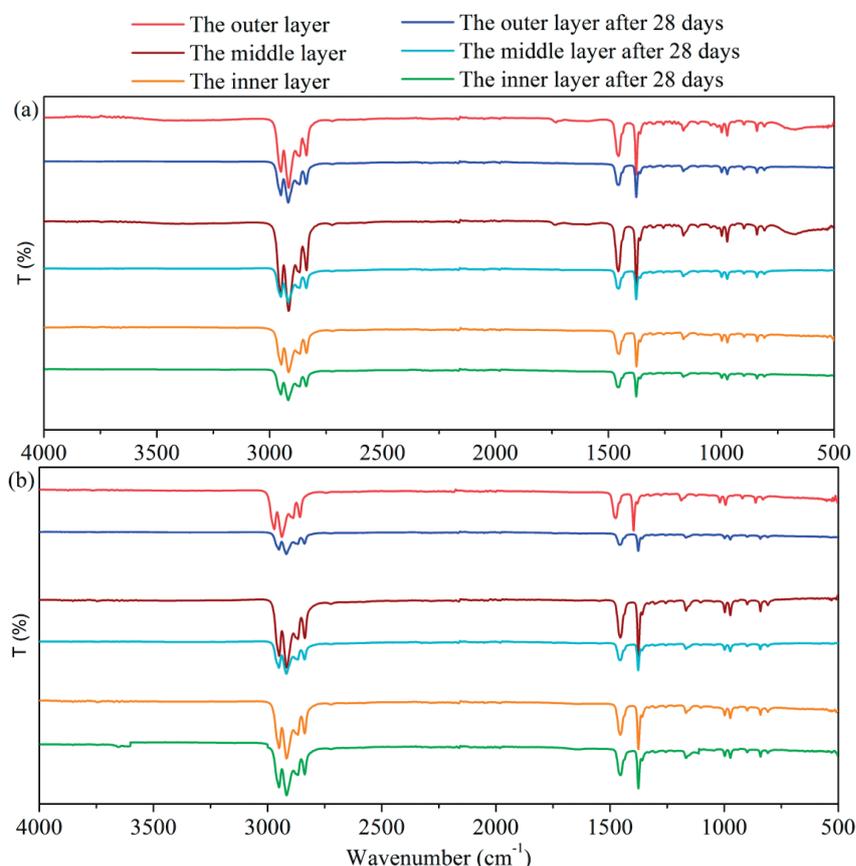


Fig. 1. Changes of infrared spectrum of N95 mask a) and surgical mask b) under UV irradiation.

produce free radical ions, resulting in the shedding of a large number of MPs [33]. The different structural changes of the two kinds of multiple use of masks after UV irradiation were mainly reflected in mask's middle layer. The intensity of each peak in the middle layer of surgical mask decreased slightly with the increase of UV irradiation time, but some small peaks appeared between 1640-1550 cm^{-1} . These small changes mean that part of the C-H bond breaks, forming a double bond structure [34]. The middle layer of surgical mask is charged, which is the most significant difference between it and N95 middle layer. UV irradiation and humid environment can easily reduce the charge content of fibers in the middle layer of surgical mask [35], resulting in the falling off of MPs.

Effect of UV Radiation on MPs Released by Masks

The mask after ultrasonic treatment was prone to aging under UV irradiation, and a large number of MPs particles fall off from N95 and surgical masks (Fig. 2a). This was consistent with the conclusion that the mask was discarded into the environment, the number of MPs increased sharply under the conditions of light, wind, rain, friction and so on [36]. The amount of MPs released by the two kinds of masks increased with the extension of UV irradiation time. It can be seen that the number of MPs falling off of surgical mask was significantly greater than that of N95 from 0-7 days (Fig. 2a). That may be due to more loose plastic fibers on the surface than N95 during the production of surgical mask, which fall off in short-term UV irradiation. This process was similar to the ultra-fine fiber discharged from the fabric in the washing process [37]. At 7-14 days, the total number of MPs produced by the two kinds of masks was basically the same, and the increase of MPs from day 7 to day 14 was not obvious. It can be seen that it takes a certain time for mask fibers to wear and degrade into MPs in the natural environment. During the period of 14-28 days, the two kinds of masks ushered in a big outbreak of MPs falling off. At this time, the number of MPs falling off N95 was significantly higher than that of surgical masks. It can be seen that after 14 days of UV irradiation, the mask fiber was worn and degraded, resulting in a large number of falling off of MPs. With the passage of time, the macroplastics falling off from the mask in the early stage may gradually decompose into MPs, which could greatly increase the number of MPs [38, 39]. Some studies have found that macroplastics have a high ability to release MPs into water [40]. From 21 to 28 days, the shedding amount of MPs of both masks decreased, previous studies have shown that mask polymers can be crosslinked with some functional groups through short-term UV irradiation, resulting in the reduction of MPs production [41]. In general, when the mask is in the environment for more than 14 days, it will produce explosive MPs pollution, and the amount

of MPs produced by N95 is higher than that of surgical mask. In order to avoid a large amount of MPs produced by the mask, the mask should be collected and treated within 14 days.

Fig. 2b) shows the color distribution of MPs produced by N95 mask and surgical mask after UV irradiation. Three kinds of MPs were detected in the MPs produced by the mask after 28 days of UV irradiation, and the transparent MPs accounted for the vast majority, about 94%-100%. This is consistent with the conclusion with Wu et al. [42] that most of the MPs falling off from different kinds of masks were transparent. This is mainly because the masks are mainly composed of colorless fibers, so the fallen MPs transparent dominated [43]. The longer the mask was in the environment, the more color MPs were produced. After 28 days of UV irradiation, the blue and red MPs of N95 mask reached 8.5% and 2.9% respectively, and the blue and red MPs of surgical mask reached 6.8% and 3.0% respectively. Other colors were detected as red and blue, and the main color was blue. Blue fabric usually appears in the outer layer of the mask, which makes them more vulnerable to radiation shedding [44]. After UV irradiation, it is first absorbed by the unsaturated bond or chromophore in the outer mask material to form the polymer free radical (unsaturated point). The free radical reaction eventually leads to the chain breaking and cross-linking of the polymer. Therefore, the outer layer with color material is the easiest to degrade and produce a large number of MPs particles. It is found that colored MPs can adsorb more harmful substances such as heavy metals and organic pollutants, thus causing more toxic effects on the organisms that like to eat colored MPs [45], thus increasing the threat to the environment.

The Particle Size of MPs Produced by UV Irradiation of Masks

Fig. 3 shows the particle size distribution of the MPs produced by UV irradiation of N95 and surgical mask. All MPs samples were divided into 6 groups according to different particle sizes. During the 28 days of the reaction, the total amount of MPS of <100 μm , 100-500 μm and 500-1000 μm accounted for more than 70% of the MPs produced by the two kinds of masks, and the highest content reached 81.83%. It can be seen that the MPs produced by the mask after UV irradiation was mainly small particle size. Moreover, after 28 days of UV irradiation, the total amount of MPs by N95 mask and surgical mask reached the highest, which were 114 particles/ cm^2 and 87 particles/ cm^2 , respectively. Researchers studying plastics such as polyethylene, polypropylene and polystyrene found that it took time for the pieces of plastic to get smaller ones [46]. Because the waste cannot be recycled in time, the discarded disposable masks accumulated in environment for a long time, which may increase the time to produce smaller MPs

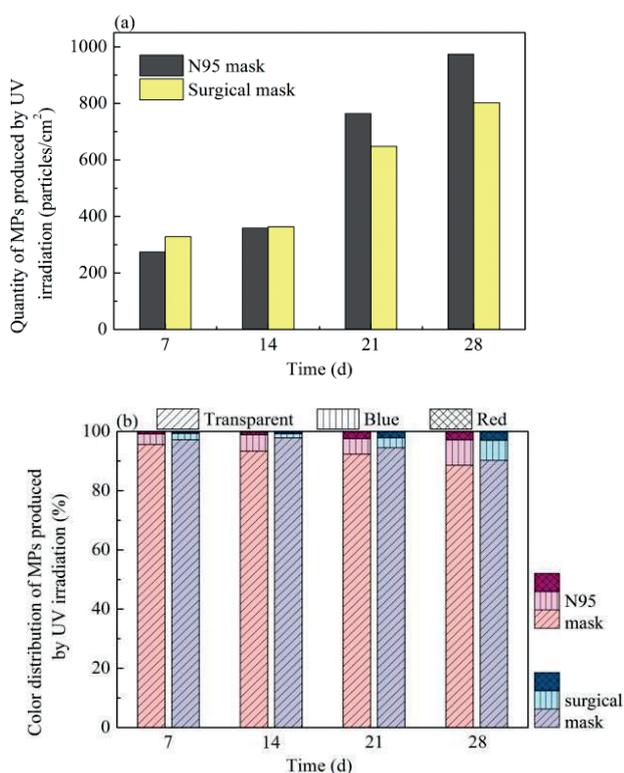


Fig. 2. Quantity a) and color distribution b) of MPs produced by UV irradiation.

particles. This explains the reason why the amount and proportion of three kinds of small-size MPs in two kinds of masks increase with time. Studies have found that small particles of MPs are difficult to be removed by sewage treatment plants, and small particles of MPs are easier to enter the organism, causing biological toxicity to cells, dissolved oxygen, etc. Therefore, a large number of discarded disposable masks into nature will inevitably lead to environmental deterioration.

It can be seen from Fig. 3 that the proportion of MPs particles <100 μm in N95 mask decreased with the

extension of reaction time, from 36.94% on the 7th day to 22.24% on the 28th day. The size of MPs particles dropped from surgical mask was slightly different from that of N95, and the proportion of MPs particles <100 μm showed a trend of first increasing and then decreasing. During the whole process, the proportion of 100-500 μm MPs of the two kinds of masks increased continuously. The proportion of N95 masks increased from 24.56% (7th day) to 38.24% (28th day), while that of surgical masks increased from 34.78% (7th day) to 42.56% (28th day). That is to say, if the dense plastic fibers on the two kinds of masks want to fall off, they need longer UV irradiation time, and they are preferentially in the form of larger particles. The MPs particles larger than 1000 μm also account for a small proportion of the MPs particles that eventually fall off. On the one hand, it may be that the UV irradiation time is insufficient; on the other hand, it may be that UV irradiation alone can not make a large number of large particle size MPs fall off [44, 47]. The MPs of 1000-1500 μm in surgical mask fell off explosively on the 7th day, and decreased significantly in the later stage. This may be due to the large particle size of the loose plastic fiber adhered to the mask in the production process.

MPs Produced by Aging of Different Layers in Mask

When the mask is exposed to UV radiation, the surface will be weathered, resulting in changes in physical and chemical properties. The surface of each layer of mask will become rougher, fragile and easier to form MPs [44, 48]. It can be seen from Fig. 4a) that for N95 mask, the concentration of MPs particles falling off the outer layer was the highest, followed by the middle layer, and the concentration of MPs particles released from the inner layer was the lowest in the whole 28 days of reaction. This result is consistent with the results of previous studies. The inner layer of N95 mask is nonwoven, which has certain comfort.

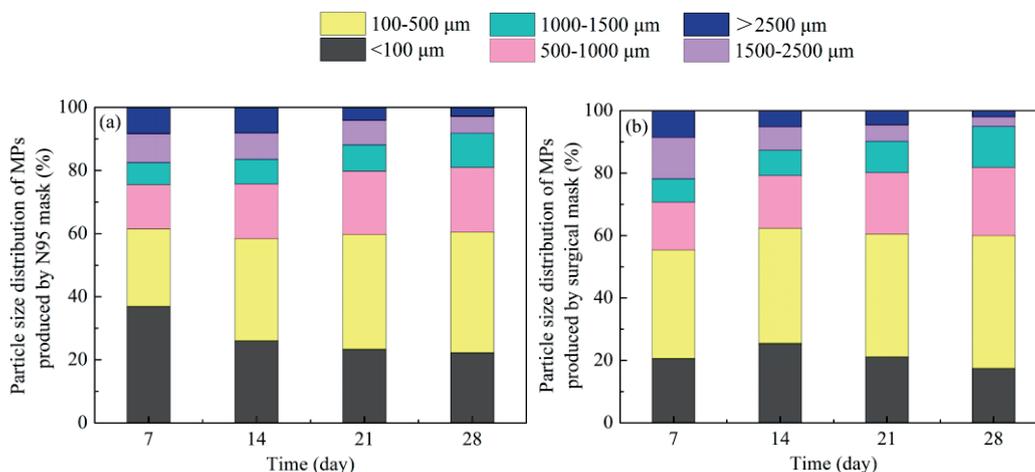


Fig. 3. Particle size distribution of MPs produced by N95 mask a) and surgical mask b).

The middle layer is made of superfine polypropylene fiber melt blown material, which can provide better filtration performance. The outer layer is nonwovens and ultra-thin polypropylene melt blown material layer, which has certain waterproof performance. The only difference between the outer layer and the inner layer of N95 mask was that the outer layer contains ultra-thin polypropylene melt blown material layer. Therefore, after 28 days of UV irradiation, the ultra-thin polypropylene melt blown material layer of MPs will fall off. The diameter of the fiber in the middle layer is about 2 μm , which is only one tenth of that of the non manufactured fabric. Therefore, the thinner middle layer is easier to shed MPs particles under UV irradiation, while the inner layer obviously falls off more slowly.

Surgical masks usually consist of three layers: the outer hydrophobic spinning adhesive layer, the middle melt blown cloth layer and the inner non-woven cloth layer. The biggest difference between N95 and surgical mask is that the filter layer in the middle of the surgical mask and the melt blown fabric used to make the mask are charge treated [35]. It can be seen from Fig. 4b) that the outer layer of surgical mask was most likely to fall off a large amount of MPs. In the 28 days of reaction, the number of MPs falling off from the outer layer was always the largest among the three layers. The second largest proportion was the inner layer. The most difficult to form MPs was the middle layer. The middle layer is a charged melt blown filter material, which will accelerate the aging of the material in the humid environment, resulting in the reduction of the charge content of the fiber in the middle layer. Therefore, there will be a certain degree of MPs falling off after UV irradiation. The outer layer of surgical mask is usually coated with polyurethane, which makes the polypropylene non-woven material have better wear resistance, good tensile resistance and less deformation. But because the main culprit of polyurethane aging is UV light, the outer mask is easy to fall off a large number of MPs particles under UV light.

Suggestions on Management of Discarded Masks

Our results show that the discarded masks without effective treatment may release a lot of MPs pollution to the environment. Therefore, suggestions on waste management was needed. UV radiation will lead to the breaking of some main chains, the reduction of molecular weight and the reduction of mechanical strength, resulting in the release of a large number of MPs. And the MPs produced by the mask are mainly small particle size. The longer the UV irradiation time, the more small particle size MPs are produced, which is more harmful to the environment. Therefore, waste masks should not be exposed to the sun for a long time after collection to prevent MPs, especially small particle size MPs, from polluting the environment. After 14 days of UV irradiation, the number of MPs produced by masks ushered in a big outbreak. Therefore, the discarded masks should be collected and disposed within 14 days as far as possible. The outer layers of N95 and surgical mask are easier to release MPs particles, and will produce more harmful colored MPs. Therefore, the outer layer of the mask should be collected and treated preferentially.

In this paper, the causes of MPs shedding of mask under UV irradiation were studied, and the particle size, color and different layer of MPs were investigated. This has guiding significance for mask shedding off MPs in the environment. However, the mask experiences wind, light, heat, wear and other factors, resulting in the shedding of MPs in natural environment. Therefore, the number of MPs falling off mask needs to be studied in practice. To sum up, future research of shedding characteristics of MPs should focus on real environment or multi factor comprehensive simulation experiment. The regulations on mask treatment should be formulated in terms of policies to avoid more MPs entering the environment.

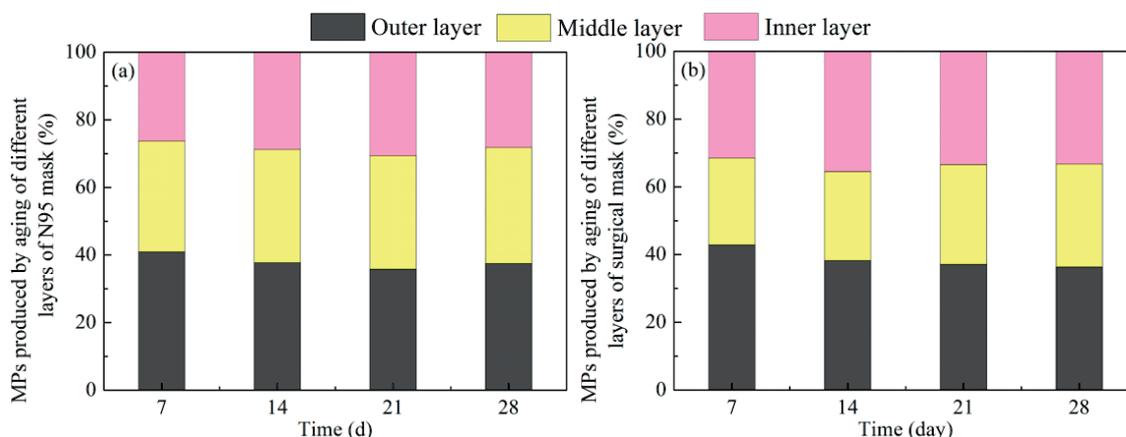


Fig. 4. MPs produced by UV irradiation on different layers of N95 mask a) and surgical mask b).

Conclusions

Due to the characteristics of rapid transmission of COVID-19 and extended survival time on the plastic surfaces, the management of masks recycling is inappropriate. Therefore, a large number of masks enter the environment without any treatment, resulting in explosion of MPs in the environment. The specific conclusions are as follows:

(1) UV radiation caused part of the main chain of the mask to break, causing a large number of MPs to release.

(2) The masks release MPs has exploded after 14 days in the environment. Therefore, discarded masks should be collected and disposed of within 14 days.

(3) The longer the UV irradiation, the more release of small particle size MPs.

(4) During the 28 days of the experiment, the middle layer of surgical mask and the inner layer of N95 mask were difficult to release MPs.

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Conflict of Interest

The authors declare no conflict of interest.

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