A two-stage cascaded ionizer for boosting PM charging in electrostatic filtration: Principles, design, and long-term performance

Yilun Gao a,b, Yuting Gu a,b, Enze Tian c,d, Jinhan Mo a,b,e,*

Department of Building Science, Tsinghua University, Beijing 100084, China
Beijing Key Laboratory of Indoor Air Quality Evaluation and Control, Beijing 100084, China
Songshan Lake Materials Laboratory, Dongguan 523808, China
State Key Laboratory for Surface Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China
Key Laboratory of Eco Planning & Green Building, Ministry of Education (Tsinghua University), Beijing 100084, China

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ABSTRACT

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In fibrous filtration for airborne particulate matter (PM), the triple wins among efficiency, resistance, and lifetime are hard to achieve owing to the filters’ intrinsic geometry. Although charging the PM has been extensively exploited, it is still far from satisfactory for practice, with drawbacks including electrode fouling, ozone, and durability. Herein, based on low-efficiency coarse filters with ultralow pressure drop and potential large dust holding capacity, we bridge this divide by introducing two-stage cascaded ionizers to boost their performance. Enabled by the separated structure for successive ion generation and transport, theoretical design and experimental results demonstrated that the ionizers could efficiently charge the PM and promote filtration efficiencies. The lab-scale prototype composed of the prepared ionizer arrays and polyester (PET) filters stably performed in 25 days, with average daily efficiency of 95.4% for 0.3–0.5 μm particles, 77.0 g/m² PM₁₀ accumulating amounts with 4.9 Pa pressure drop, and 56.2 W/m² power consumption at 1.4 m/s filtration velocity. As the “efficiency boosters” for filters, the ionizers are meanwhile free of PM contamination and possess ultralow ozone generation. We anticipate that our approach will be an efficient, durable, and energy-effective solution in the air filtration community in ventilation systems for clean and sustainable built environments.

1. Introduction

Air pollution is costly, and the status is worrisome. Despite the sharpening focus on reducing pollution, ambient particulate matter (PM) is still the leading cause of various diseases [1], including but not limited to cognitive disorders, respiratory and cardiovascular diseases [2–4], contributing to more than 0.8 million premature deaths annually [5]. In 2021, the World Health Organization (WHO) launched the updated air quality guideline for lowering the recommended limit value for PM₂.₅ (fine particles with aerodynamic diameters of 2.5 μm or less) to 5 μg/m³ [6], providing clear evidence for the damage air pollution inflicts on human health. More to the point, the association between disease mortality and long-term exposure to PM₂.₅ can even persist at low concentrations [7]. In modern society, most people spend at least 80% of their lifetime indoors [8]. As outdoor PM can penetrate building envelopes and deposit on indoor surfaces, efficient PM removal technologies have emerged as indispensable demands in heating, ventilation, and air conditioning (HVAC) systems for constructing sustainable and healthy environments.

Generally, straightforward and efficient removal of airborne PM relies on utilizing fibrous filters to block the motion of PM suspending in the air passively. PM will escape from the airstream and attach to the fiber surface, elucidated by the combined mechanically-dependent effect of Brownian diffusion, inertial impaction, and interception [9]. According to the basic filtration theory, as the airflow velocity increases, the resistance across the filters will accordingly rise, and PM will less possibly be captured due to the weakened mechanical effect. As a result, in the design of filters, improving performance is the consequence of strengthening mechanical effect by blocking airways, which increases the air resistance and decreases the dust loading capacity as an adverse impact. Distinguished from filtering facepiece respirators (FFR) for personal protection, air filters in ventilation systems always serve at high airflow rates (m/s level) and large amounts of PM loadings [10], so relieving the efficiency-resistance-lifetime trade-off is challenging yet...
critical. Utilizing the electrostatic effect has been a gold rush to strengthen PM capture in recent years. Considering that the efficiency-resistance-lifetime trade-off emerges from the intrinsic geometry of filters, electrostatic reinforcement may well be the solution to increase PM attachment efficiency at 1 m/s filtration velocity, emerging the so-called “electrical responsiveness” for the fibrous filter medium [27]. Still, the ozone generation from the corona discharge cannot be overlooked although the modified PET fibers could efficiently remove part of them.

In these electrostatic filtration devices, the PM charging units engine the performance. Given the status quo, the mentioned coronadischarged ionizers for PM charging are commonly used and proved effective, though it still faces issues to consider. Firstly, as the ionizers are exposed to polluted airflow to provide charges, with the long-term particle accumulation on the electrodes, the charging characteristics will deteriorate, at least emerging sharp increase of charging currents [28,29], excessively high energy consumption [30] and ozone generation [27]. In recent works, the tested long-term working period for ionizer-assisted filters was typically less than 300 h or just several minutes [31,32], far from the requirement in ventilation systems. Further, the PM contamination will inevitably shield the discharge, thus weakening PM charging in the filtration. To achieve higher efficiency, lifting applied voltages and reducing gaps between electrodes to approach the corona threshold will strengthen PM charging but still cause air breakdown and electro spark as a compromise [33]. More significantly, despite using PM prechargers, most of these works are based on medium-high efficiency filters, which still possess relatively high pressure drop. Considering the potential low pressure drop for coarse filters, the efforts provided by PM charging are not fully utilized.

Herein, approaching high-efficiency PM filtration technology with minimal pressure drop and maximum long-term durability, we designed two-stage cascaded ionizers to boost the efficiency of coarse polyester (PET) fibrous filters. After delineating the principles of corona charging, the key point of our strategy is the separated structure for successive ion generation and transport, which contributes to efficient PM charging, resistance to PM fouling, and low ozone generation. To confirm the feasibility of our concept, we also established a lab-scale filtration system equipped with cascaded ionizer arrays and monitored the long-term filtration performance of the filters for up to 25 days.

2. Methods

2.1. Assembly of the ionizers

Three self-made ionizers were investigated in this work: the metal pin-to-ring electrodes (MP), the cascaded metal pin-to-ring electrodes (c-MP), and the cascaded carbon brush-to-ring electrodes (c-CB).

Design of MP ionizers. In MP units, the corona discharge was driven by a high voltage direct current (HVDC) power supply (P30, Boer Co., Ltd., Suqian, China) with adjustable voltage outputs of 0 ≈ 30 kV, between a 20-mm length tungsten pin electrode (ELCT2-20A, Fujikura Ltd., Tokyo, Japan) and a circle-pored iron ring with 50-mm outer diameter. The distance between the pin tip and the centre of the ring was 10 mm.

Design of c-MP and c-CB ionizers. The photographs of the cascaded ionizers showing the ion-generating part are displayed in Supplementary Fig. S1. In the cascaded charging units, two HVDC power supplies served for separated two stages: DC-1 for ion generation and DC-2 for ion transport. High voltages were only applied in DC-1, and DC-2 was connected to display the induced voltages and currents in the circuit. The gaps between each electrode (tip of the 20-mm metal pin/carbon brush, the centre of the circle-pored ring, followed by a piece of 100 × 100 mm² copper mesh) were 10 mm and 100 mm, respectively. Both of the HVDC supplies were connected to the grounded copper meshes.

Electrostatic simulation. The corona charging field and potential differences of MP and c-MP ionizers were solved using a finite element software solver, Ansys Electromagnetics Suite 19.2. The charging voltages applied on the pin electrode and grounded plate were set at +15 kV and 0 kV, respectively. The geometry referred to the actual module described above.

2.2. Performance measurements for the ionizers

Measurement and observation of the charging characteristics. The ionic strength, \( N_i \), was detected using an air ion counter (AIC-2000, Alphalab Inc., USA) at different testing points to investigate the charging capacity of ionizers. The tip of the pin/carbon brush, the centre of the circle-pored metal ring, and the testing point were on the same line. Besides, the electric corona was observed by using an in-situ time-lapse mobile camera (P30 Pro, Huawei Technology Co. Ltd., China). The images were collected for continuous 60 s to obtain each optical photograph.

Testing the initial filtration performance. The filtration test was conducted in a customized acrylic duct with a cross-section of 80 mm × 80 mm, and the length between the charging module and the filter was 100 mm. The single ionizer was installed in the duct for PM precharging to improve the filtration performance. We used \( U_0 \) to denote the voltage applied to the ionizers. The ambient particles were driven through the duct after a driven fan, a steady flow plate, the PM precharging units, and a piece of commercially available 8-mm-thick PET fibrous filter. A peristaltic pump (550D, Fujiwara Co. Ltd, China) was used to supply 1 L/min clean air to avoid PM fouling on the ionizers. The airflow velocity in the duct can be adjusted by the fan and set at 1.0 m/s. The filtration test was conducted indoors at 19.7–26.8 °C, with a relative humidity of 17.9%–39.4%. Before every single experiment, the inner surface of the duct will be washed and cleaned with alcohol wipes to prevent charge accumulations. The 8-mm-thick coarse PET filter with large fiber distance, fluffy structure, and commercial accessibility was selected as a representative. The received PET pieces were pre-treated by immersing in isopropanol for 5 min and drying in an oven at 75 °C for 2 h to remove residue charges. Before every single experiment, a new piece would be
The images of PET filters were captured by a field emission scanning electron microscope (FE-SEM, Merlin VP compact, Carl Zeiss AG), as shown in Supplementary Fig. S2. The detailed structure of the experimental setup is available in the schematic Supplementary Fig. S3.

The number concentrations of 0.3–5 μm particles were measured by the optical particle counters (Aerotrak 9306, TSI Inc., Shoreview, USA) at both upstream and downstream of the filter. Besides, the number concentrations of 11.5–273.8 μm particles were measured by a scanning mobility particle sizer (SMPS 3910, TSI Inc., USA). The particle counts were recorded every 1 min for a single sampling. The removal filtration efficiency η of particles for one observation with a specific size of dp (μm) was calculated by Eq. (1).

\[
\eta(d_p) = \left( \frac{C_{up}(d_p) - C_{down}(d_p)}{C_{up}(d_p)} \right) \times 100\%
\]

where, \(C_{up}(d_p)\) and \(C_{down}(d_p)\) are the particle counts (#/cm\(^3\)) of a specific diameter of \(d_p\) (μm) at the upstream and downstream of the filter. If not specifically mentioned, the single efficiency value was calculated by averaging \(C_{up}(d_p)\) and \(C_{down}(d_p)\) from three single samplings. We used the symbol \(\eta_{0.3}, \eta_{0.5}, \eta_{1.0}\) and \(\eta_{3.0}\) to denote \(\eta(d_p = 0.3–0.5 \mu m)\), \(\eta(d_p = 0.5–1.0 \mu m)\), \(\eta(d_p = 1.0–3.0 \mu m)\) and \(\eta(d_p = 3.0–5.0 \mu m)\), respectively.

Besides, the pressure drop across the filter was measured by a differential gauge (DP-CALC 5825, TSI Inc., USA). The filtration face velocity was measured by a thermo-anemometer (435-1, Testo SE & Co. KGaA, Germany) at the air duct exhaust.

### 2.3. Long-term filtration monitoring

The filtration prototype. We established a tailor-made lab-scale filtration prototype (schematic Fig. 1(a)) equipped with c-CB arrays for PM charging and folded electrostatic PET filters for PM collecting. In brief, the single c-CB ionizers were assembled in an array and packaged as modules for installation in the prototype. Along with the assistance of c-CB ionizers, we applied an electric field to the coarse PET filters to collect charged PM for maximizing the electrostatic filtration effect according to the methods in previous research [34]. NaCl solution (0.9 wt %) was filled in the nebulizer for stable PM generation, and a peristaltic pump (550D, Fujiwara Co. Ltd, China) was used to supply 3 L/min clean air for PM feeding. The detailed assembly of the prototype is shown in Supplementary Fig. S4. The detailed design, structure, and operating conditions of the prototype were described in the Supplementary Information.

**Measurements and calculations.** The efficiency \(\eta_{0.3}\), pressure drop, ozone generation, accumulated PM\(_{10}\) collecting amounts \(M_{acc}\), and energy consumption were monitored and calculated in this study to evaluate the long-term performance of the prototype. The measurements of the first two parameters are illustrated above in Section 2.2. The ozone generation of the prototype (ppb, part per billion) was obtained using an ozone monitor (Model 205, 2B Tech., USA) every 12 h for five continuous samplings. The accumulated PM\(_{10}\) amounts were approximately estimated by a similar method in the previous study [35], as shown in Eqs. (2) and (3).

\[
M_{acc, \text{PM}_{10}} = \sum_{d=1}^{n} m_{\text{PM}_{10}} \cdot \text{d} \cdot \text{t}
\]

where, \(M_{acc, \text{PM}_{10}}\) is accumulated PM amount after \(n\) day(s), g/m\(^2\); \(m_{\text{PM}_{10}}\) is PM daily collecting amount in the \(d\)\(^{th}\) day, g/m\(^2\); \(\rho\) is the density of NaCl particles, 2.16 g/cm\(^3\); \(v_{\text{air}}\) is the air velocity in the testing air duct, m/s; \(Q_{s}\) is the sampling air amount of the particle counter per minute, 2.1225 L; \(t\) is the time in a day, minute; \(d_{dp} = 1–6\) represent particle sizes of 0.3, 0.5, 1, 3, 5, and 10 μm, respectively.

The accumulated energy consumption \(W_{acc}\) (kWh/m\(^2\)) of the whole device was calculated by Eq. (4).

\[
W_{acc} = \sum_{d=1}^{n} \left( 24 \times \frac{P_{l,d}}{1000} \right)
\]

where, \(W_{acc}\) is accumulative energy consumption after \(n\) day(s), kWh/m\(^2\); \(P_{l,d}\) is the power use per unit filtration area in the \(d\)\(^{th}\) day, W/m\(^2\).

### 3. Results and discussion

#### 3.1. Structure design for the ionizers

In fibrous filtration, the airflow-carried PM will bypass the fibers, and be captured according to the combined effect of Brownian diffusion, interception, and inertial impaction [9]. These mechanically-independent effects factor in the inherent efficiency-pressure drop conflict for fibrous filtration.

Supposing the PM carries sufficient charges. With the electrostatic effect, when PM moves to the fibers, the PM-fiber interaction occurs as the ion-induced effect at this moment and can be defined as image force, \(F_{i}\), as shown in Eq. (5).

\[
F_{i} = \frac{1}{16\pi\varepsilon_{0}} \frac{\varepsilon_{i} - 1}{\varepsilon_{i} + 1} \frac{q^{2}}{(r - \frac{d}{2})^{2}}
\]

where, \(\varepsilon_{0}\) is the vacuum dielectric constant, 8.854 \(\times\) 10\(^{-12}\) F/m; \(\varepsilon_{i}\) is the
relative dielectric constant of the fiber; \( q \) is the charges on the particle (C); \( r \) is the radial distance from the fiber centre to the particle (m). For coarse filters with large fiber diameter and fiber distance, charging the PM can initiate PM-fiber electrostatic interaction without blocking airways, thus being the pushing hand for improving PM collecting efficiency. By singly charging the PM with \( \sim 0.3 \mu \text{m} \) diameters, the efficiency increase of "electrically-responsive" coarse filters with specially-tuned surface morphologies and electric properties can be even as large as \( >50\% \) [10].

A traditional corona-charging ionizer usually consists of two electrodes with different curvatures: one is the spiculate discharging electrode, and another is to be the grounded plate. As direct current high voltages are applied between them, a non-uniform electric field will generate. If the field intensity exceeds an onset threshold to initiate corona, the air nearby will be ionized into ions and electrons [36]. Then, these two-fold charges will be accelerated by the electric field, and part of them (depending on the voltage polarity) will head to the ground and collide with the suspended airborne PM carried by the airflow. Further, the charged PM will aggregate and deposit on the collecting filters for electrostatic filtration. The actual corona onset field intensity between the pin-to-ground electrode, \( E_{\text{onset}} \) (kV/cm), can be empirically represented by Peek’s Law in Eq. (6) [37].

\[
E_{\text{onset}} = E_0 \left( 1 + \frac{K}{(\frac{r}{\delta r})} \right) -\delta m \tag{6}
\]

where, \( E_0 = 31 \text{ kV/cm} \) is the standard onset field intensity in the air; \( m \), a dimensionless coefficient, indicates the smoothness of the discharging electrode surface; \( K = 0.308 \text{ cm}^2 \text{g}^{-1} \), \( \delta \) is the relative air density compared to that at the standard air temperature (25 °C) and atmospheric pressure (101.325 kPa); \( r \) (cm) is the radius of curvature of the pin electrode. Thus, the onset electric field threshold can be reached by applying \( U_c \) in Eq. (7).

\[
U_c = \frac{r}{2} E_0 \ln \left( \frac{r + 2d}{r} \right) \tag{7}
\]

where, \( U_c \) is the corona onset voltage (kV), and \( d \) (cm) is the electrode distance.

For efficient PM charging, the ionizers should generate abundant charges; For another, as airborne particles abound in the polluted airflow, the evenness of the corona charging field will also contribute. Once the applied voltage on the electrodes is enhanced to approach the threshold for chasing extreme charging efficiency, it takes risks to form air breakdown and generate a mass of ozone in a second. Moreover, in the MP ionizers shown in Fig. 2a, the electric field originates from the pin tip locally. As the electric field line only originated from the pinpoint and ended up at the ring, the uneven field distribution may be incompetent for PM charging when aerosol particles are filled in the airflow.

The schematic of the c-MP ionizers is shown in Fig. 2b. The c-MP ionizer includes three electrodes: a tungsten pin for initiating corona, a relaying metal ring for generating an induced electric field, and the grounded metal mesh. Similarly, high voltages can be applied between the pin and the grounded electrodes, and the charging field first forms between the pin tip and the relaying metal ring (electrodynamic Stage 1 for ion generation). Thus, the ions will be generated in this zone through the corona charging between the pin tip and the relaying ring. In Fig. 2b, as the relaying electrode is not connected to the ground, \( \delta U_1 \) will be smaller than \( U_c \) in c-MP units, while they are equal in MP units. Therefore, at the same \( U_c \), the strength of the charging field for cascaded ionizers is weaker than that in MP units (see the difference in schematic Fig. 2a and b). In other words, to generate more ions between the 10-mm gap, a larger \( U_c \) can be loaded within tolerable limits in c-MP units to reach the breakdown threshold, and the risk of generating ozone can be reduced. Then, in Stage 2, the relaying metal ring with a high electric difference combined with the grounded mesh will act as the electrodes. When applying \( U_c \) in Stage 1 to create \( \delta U_1 \), an induced potential difference will emerge in Stage 2 (\( \delta U_2 \)), and \( \delta U_2 \) will increase as \( \delta U_1 \) goes

Fig. 2. Schematics showing the structures of a MP and b c-MP ionizers. The simulated corona charging fields of c MP and d c-MP ionizers, showing the field distribution differences.
up. Supplementary Fig. S5 shows the voltage correspondence of the charging and induced circuit in cascaded electrodes. It is noteworthy that as no voltage was applied, the field in Stage 2 was spontaneously induced from the charging field in Stage 1. As both electrodes are planar in shape, unlike the divergent charging field generated from the pin tip in Stage 1, the field in electrostatic Stage 2 for ion transport will uniformly distribute, as demonstrated by the simulated schematic in Fig. 2c and d. In all, the generated ions will be accelerated, delivered to the grounded mesh, and finally received by the suspended PM in the airflow. In this manner, the two-stage cascaded ionizers can generate ions in series, form “ionic curtains”, and efficiently charge airborne PM ultimately.

Besides, the charging pin in the aforementioned c-MP electrodes can be replaced with carbon brushes (CB) for further investigation. The charging characteristics for the c-MP and c-CB ionizers will be discussed in the following part.

3.2. Measurement for the ionic strength

Testing the ionic strength can directly reveal the charging performance of the ionizers. The generated ionic concentrations in the airflow, \( N_i \), were measured to verify the design of the ionizers. While the tip of the pin/carbon brush, the centre of the metal ring, and the sampler were in order on the same line, we denoted the sampling distance as the length from the ring to the sampler in Stage 2. Theoretically, \( N_i \) will be larger when the sampler gets closer to the electrode and when \( U_c \) increases. Fig. 3a presents the ionic concentrations for three ionizers at different distances at 9-kV applied voltage. With the sampling distance decreasing from 10 cm to 2 cm, more ions were detected as the charging field originated from Stage 1. Specifically, at a 2-cm sampling distance, the detected ionic numbers were 55.7 × 10^6, 98.7 × 10^6, and 156.0 × 10^6 cm\(^{-3}\) for MP, c-MP, and c-CB ionizers, respectively. Due to the induced field performing in cascaded ionizers, more ions were transported to Stage 2 and detected, which aligns with our design principle. In addition, as shown in Fig. 3b, the ionic countings at 9 kV increased to 40.1 × 10^6 cm\(^{-3}\) when installing carbon brushes, indicating that applying carbon brushes would further promote ionic concentration.

Fig. 3c and d performed ionic strength variations with different \( U_c \) for c-MP and c-CB ionizers. With the adjustment of \( U_c \) from 7 to 9 kV, the ionic signal was not evident, while the voltage was insufficient to trigger the onset corona for charging. Then, the values significantly rose when \( U_c \) was upregulated from 9 kV to 15 kV, showing sufficient ions were generated for the developed cascaded ionizers. When \( U_c \) was as high as 15 kV, although all the sampling distances were monitored, with less than 4-cm distance, the ion countings were in large quantities and even beyond the rangeability of the ionic counter (see the scatters marked with *). Supplementary Fig. S5 also demonstrated that the induced \( \delta U_2 \) in c-CB ionizers was larger than that in c-MP, which supports our outcomes. Thus, it concludes that the c-CB and c-MP ionizers can boost PM charging, and c-CB will be more effective.

The electric arc will be initiated and stably exist at the electrode’s tip during corona discharge. Still, the arc triggered by direct current (DC) high voltage will be hard to observe in natural lighting. Here we constructed an in-situ observation platform using a time-lapse camera to record the characteristics of cascaded ionizers in the dark room, as displayed in Fig. 4a. Generally, the DC arc in the air emerges blue and purple, and the arc’s shape can reflect the strength and distribution of the charging field. In the c-MP electrodes, the arc was concentrated at the pin tip and went straight down to the grounded electrodes. With the growing \( U_c \) from 9 to 15 kV, the color turned purple to white (Fig. 4b-e), resulting from the fortified charging field and energy output. For comparison, the arc from c-CB electrodes was not recorded by the lens but showed scattered, dispersive light spots at the brushing tips locally (Fig. 4f-i). Notably, given that at the same volume, more brush hairs will exist in the charging unit than the single metal pin, the energy intensity from the electric field will be shared, and the charging field in the c-CB ionizers will be more uniform-distributed for ion generation and transport.

3.3. Initial filtration performance

Fig. 5a–d presents the filtration efficiency of the ionizer-assisted PET filter at different \( U_c \). In general, the efficiency augments with the particle diameter increasing, which is in line with the reported results towards electrostatic filtration [38]. With the adjustment of \( U_c \) from 9 to 15 kV, the PET filter performed enhanced filtration efficiency attributed to the improved charging performance of cascaded ionizers. For instance, when equipped with c-MP ionizers, the \( \eta_{0.3} \) at 1 m/s filtration velocity was steadily enhanced from 31.8%, 45.9%, 58.2% to 65.0% at 9, 11, 13, and 15 kV applied voltages, respectively. Considering that the pristine coarse PET filters performed an initial \( \eta_{0.3} \) of only 4.9%, the utilization of cascaded ionizers considerably expanded their capacity for capturing particles by efficiently charging them. That is, the cascaded ionizers act as “efficiency boosters” to overcome the intrinsic trade-off for fibrous filtration, especially for those coarse filters with poor initial filtration capacity and intrinsically ultralow pressure drop.

Moreover, the cascaded structure in these ionizers possessed natural advantages for a broader range of applying voltages. As the potential difference between electrodes is lower in MP ionizer, it reflects in Supplementary Fig. S6 that when \( U_c \) was upregulated to 10 kV, the overcharged electric field led to air breakdown, and the module could not operate stably to provide effective PM charging, so the filtration efficiency would diminish. Meantime, the ozone generation reached 101.7 ppb with acute discharge breakdown.

In the cascaded ionizers, we further prove that carbon brushes can be an effective alternative to traditional metal pin electrodes. Fig. 5a-d displayed that when it came to c-CB ionizers, \( \eta_{0.3} \) increased to 38.3% at 9 kV and 68.8% at 15 kV and were slightly higher than that using c-MP ionizers. Noticing that the ion countings generated from c-CB ionizers were detected to be larger in Section 3.2, it can be speculated that more particles can be efficiently charged, and the fortified electrostatic PM-fiber interaction resulted in improved outcomes. Moreover, carbon brushes provided more charging tips than the single metal pin, also
performing a uniform electric field in Stage 1 for ion generation.

In MP ionizers, as the charging units expose to airflow and the passing PM attaches to the electrodes, the charging characteristics will be deteriorated by the contamination, and the filtration efficiency will decline [30]. In addition to the qualified initial performance, the durability of the single cascaded ionizers is examined by continuous experiments up to 168 h when $U_c$ was fixed at 15 kV. The filtration performances up to 168 h for c-CB equipped PET filters are displayed in Fig. 6a. Using the ambient PM source introduced from outdoors, the average $\eta_{0.3}$ per 12 h for the c-CB PET filter reached 72.1% with slight variations. In cascaded ionizers, the ions are generated by charging in Stage 1, and PM will exchange with ions and be charged in Stage 2. Thus, the PM contamination of charging electrodes can be avoided. Further, in Fig. 6b, the average $\eta_{0.5}$, $\eta_{1.0}$ and $\eta_{3.0}$ for c-CB ionizers increased stepwise to 75.8 ± 2.2%, 84.5 ± 1.9% and 86.8 ± 3.7%, respectively. Besides, as summarized in Fig. 6c and Supplementary Fig. S7, the c-MP assisted PET filter also performed slightly lower but stable efficiencies in the test. Therefore, these steady removal efficiencies revealed their durable charging properties.

3.4. Long-term filtration performance

In real applications, the filtration performance is expected to sustain continuous particle accumulations for at least several weeks, and the PM charging unit should be available for actual use. For easy comparison, we experimented for 25 days until the filters collected comparable PM amounts with other reported works. The concentrations of the generated PM source are provided in Supplementary Fig. S8. As displayed in Fig. 7, the long-term daily $\eta_{0.3}$, $\eta_{0.5}$, $\eta_{1.0}$, and $\eta_{3.0}$ remained at high levels of averaging 95.4%, 96.7%, 97.8%, and 98.1%, respectively. Given that the efficiencies would vary in a narrow range owing to the fluctuation of environmental parameters and PM concentrations [30], the c-CB arrays can continuously be well-operated on the strength of their stable charging performance under disturbance in actual situations. Besides, applying charging arrays can further prolong the exposure time between PM and ions, thus promoting charging effectiveness and evenness. In addition, the efficient c-CB ionizers also contribute to multiplying the filtration for coarse PET filters with very low additional energy costs. As shown in Supplementary Fig. S2, PET filters with 8-mm-thickness in this work possessed only 159 g/m² gram weight, ~30 μm averaging fiber diameter, and most importantly, ~300 μm fiber gaps, which is
nearly 100 \times larger than the submicron PM diameter. These filters showed superior permeability for lowering energy consumption, causing low PM capture efficiency as well. As c-CB ionizers act as "efficiency boosters" for coarse filters, we monitored the accompanying "costs" of the whole filtration prototype, including electric dissipation and the fan consumption used to overcome filters' air resistance. After 25 days, the PM$_{2.5}$ and PM$_{10}$ collecting amounts for PET filters steadily accumulated to 35.9 g/m$^2$ and 77.0 g/m$^2$ (see in Fig. 8a), while the pressure drop increased from initially 2.5 Pa to 4.9 Pa (see in Fig. 8b). As the charged particles be effectively captured and attached on the fibers during the experiment, the pressure drop lifting of the whole filtration prototype would be the increase of filters' air resistance. On the other hand, the electric dissipation would be decided by the running characteristics of c-CB arrays. At the pressure drop of 4.9 Pa and PM$_{10}$ collecting amount of 77.0 g/m$^2$, as presented in Fig. 8c, the accumulated energy consumption of the whole module reached 33.7 kWh/m$^2$ filtration area, which can be converted into 1.3 kWh/m$^2$ per day and 56.2 W/m$^2$. Although high voltages were supplied, as shown in Supplementary Table S1, c-CB arrays performed the steady charging currents (0.020–0.040 mA) and ultralow ozone generation (below 2.0 ppb) during the 25-day running period with ever-changing environmental and operating parameters, exhibiting their durability, safety, and insensitivity when being available in actual situations. It should also be emphasized that given the separated structure for ion generation and PM charging, the cascaded ionizers can defend themselves against pin contamination from PM deposition, and the charging characteristics can perform steadily and be effective.

### 3.5. Comparison with other studies

Compared to the traditional pin-to-plate ionizers, the structure of cascaded ionizers contributed to electrostatic filtration by achieving relatively high efficiency, stable charging characteristics, and low energy consumption. Firstly, in Fig. 9a, we compared the overall long-term efficiencies of PET filters assisted by c-CB ionizers and fore-cited MP ionizers [30]. In the period, the daily $\eta_{0.3}$ in the reference varied from 67.6% to 95.2%, with an average of 86.6%, significantly lower than the reported value in this work. Notably, as the charging pins were exposed to the airflow in MP units, the corona discharge was shielded with the non-conducting PM accumulating on and covering the electrodes. Thus, the weakened strength for PM charging resulted in efficiency decay for the MP-assisted prototype after approximately 15 days, while the performance for the c-CB prototype remained at high levels. In addition, we made a comprehensive comparison in Fig. 9b towards the range of charging currents, pressure drops, and accumulated energy consumption between these two prototypes. Overall, the cascaded modules possessed decreased pressure drop and power consumption with higher and more stable efficiency. Namely, in MP modules, it was proved that the energy consumption of the filtration setup had a significant positive correlation with the PM collecting amount. Specifically, while the charging currents for MP units fluctuated from 0.045 to 0.156 mA after 25 days to lift the charging consumption, the currents of c-CB ionizers maintained at 0.020–0.040 mA.

Unlike the filters manufactured for face masks or window screens, the filtration prototypes for ventilation systems always work at high
filtration velocities (m/s level), though simultaneous high filtration efficiency and low pressure drop can hardly achieve. As filtration performance is only comparable when the devices are tested at specific filtration velocities and for PM with similar size, we compared these performances of the c-CB prototype with other reported technologies utilizing PM charging. The efficiency-pressure drop trade-off for these techniques is illustrated in Fig. 10. In Supplementary Table S2, we also listed all the detailed data from comparable candidates, including their filter materials, efficiencies, pressure drops, filtration velocities, and energy consumptions. Overall, the c-CB PET filters showed ultralow initial pressure drop (2.5 Pa) at high filtration velocity (1.4 m/s) and excellent PM capture efficiency among all these candidates but possessed relatively low power consumption. The stable efficiencies and controllable pressure drop increase to 4.9 Pa after 25 days also supported the durability of our prototype. One reason for this may be the geometry of PET filters. Ascribed to the large fiber distance and low fibrous solidity, the coarse PET substrates provide higher permeability, especially under large air velocities than those technologies based on medium-high air filters. More significantly, the full potential of coarse filters can be reached by applying cascaded ionizers, and the cost of electric dissipation is limited. As the sufficient and stable PM pre-charging supplied by cascaded ionizers, the efficiency and long-term usability for coarse filters can be dramatically fortified, thus achieving triple wins for overcoming the intrinsic compromise between efficiency and pressure drop under large operating airflow velocities.

3.6. Application and perspectives

As the basic operating principles and a case prototype were proposed above, deeper investigations can be conducted for pushing the real applications of cascaded ionizers. Firstly, the charging strength herein was quantitatively evaluated by monitoring the charging voltages, currents, and ionic strength. Although filtration efficiency is the gold standard for evaluating the charging performance of cascaded ionizers, directly revealing the PM fractional charge amount decided by the charging mechanisms should be of equal importance. Thereafter, the performance for removing particles covering nano and submicron sizes can be obtained and estimated comprehensively.
Further, as illustrated in Section 3.1, since the charging characteristics are highly correlated to electrodes' shapes and materials, the structure of ionizers can also be optimized for applications. Adjusting the gaps between electrodes will also contribute. Substantially, charging the PM is a mass transfer process relying on the interaction between the electric field and the flow field. Herein, although the flow distribution of cascaded ionizer arrays can ensure efficient PM removal, improvement can still be put into effect. For example, establishing a multilevel structure or distributing the airflow to small streams can enhance ionic transfer, and the efficiency of the filtration unit can be further boosted. Lastly, nebulized NaCl was used as the target PM to monitor long-term transfer, and the efficiency of the filtration unit can be further boosted.

4. Conclusions

Efficiency-pressure drop trade-off is ubiquitous in fibrous filtration. Whilst many routes were followed to endow airborne PM with charges for lifting electrostatic filtration effect, this work designed the two-stage cascaded ionizer for boosting the performance of coarse filters, which originally possessed inherent low pressure drop and poor initial efficiency. We delineated the design and principles for the cascaded ionizers, and confirmed their properties by quantitative ionic measurement, in situ time-lapse observation, and PM filtration monitoring. As the ions were generated, transported, and exchanged with PM in successive and synergistic two stages, the cascaded ionizers can effectively perform with high filtration synergy, durability, and low energy consumption. In the tailor-made lab-scale prototype equipped with the c-CE ionizers, an electrostatic 8-mm-thick coarse PET filter showed improved 0.3-0.5 μm PM removal efficiency of averaging 95.4% for 25 days and ultralow pressure drop of 2.5 Pa, collecting 77.0 g/m² PM10 to achieve large holding capacity with the cost of only 56.5 W/m² consumption. Considering the initial 4.9% efficiency for the pristine filters, the improvement was significant. Compared with the traditional corona-charging ionizers, the ionic counts generated by cascaded ionizers increased 1.8 times with nearly no oxygen generation (lower than 2.0 ppb). As a cleaner technology to minimize hazardous airborne PM, we anticipate that our purification approach will provide valuable insights into developing energy-efficient air-cleaning devices, thus contributing to a healthy and sustainable environment.

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Appendix A. Supplementary data

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References
