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Plasma jets effectively promoting cloud condensation nuclei formation and aerosol activation in artificial weather modification

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ABSTRACT

Enhancing precipitation is of paramount importance in water-scarce regions. The availability of cloud condensation nuclei (CCN) is a critical prerequisite for precipitation formation. This study investigates a novel approach to promoting CCN formation by using plasma jets. The results indicate that after 30 min of plasma jet operating, the CCN concentration increases by a factor of 1.1–2.6, while the aerosol activation rate increases by approximately 2.5 times. The concentration of CCN remains high even after the plasma jet is turned off, which is attributed to the significant number of electrons and ions generated by the plasma jet, facilitating aerosol charging. The main mechanisms driving the formation of CCN are ion-ion recombination and reduced free energy barrier during water vapor condensation following aerosol charging. These experimental findings demonstrate the feasibility of using plasma jets to enhance the formation of CCN, with significant implications for artificial weather modification, particularly in the context of climate change.

I. INTRODUCTION

Water scarcity is a pressing issue affecting many countries and regions globally.[1] Artificial precipitation enhancement holds immense potential in addressing this challenge.[2–4] Sufficient number of cloud condensation nuclei (CCN) are the basis for cloud formation and precipitation.[5,6] However, conventional methods of seeding hygroscopic salts as CCN used in warm cloud catalysis suffer from limitations such as high costs, storage complexities, and environmental pollution, hindering the progress of warm cloud catalyzed rainfall. Consequently, there is a critical need to develop an efficient, economical, and environmentally friendly method for increasing CCN in warm clouds. Traditional warm cloud catalysis techniques primarily focus on introducing CCN directly into warm clouds, often overlooking the contribution of atmospheric aerosols. Recent studies have shown that atmospheric aerosols, when properly activated, can serve as effective CCN, offering a more sustainable alternative to traditional seeding methods.[7,8] Several scholars have carried out applications of plasma-activated aerosols in the environment, agriculture, and medicine. It was shown that plasma-aerosol interactions are highly efficient and environmentally friendly, but their application in weather modulation still needs further research.[9–14] For instance, laboratory experiments have demonstrated that ions and charged particles can significantly enhance the activation of aerosols into CCN, particularly under controlled conditions.[7,12] This suggests that leveraging existing atmospheric aerosols, rather than relying solely on external seeding agents, could provide a more efficient pathway for cloud formation and precipitation enhancement. By utilizing the natural abundance of aerosols in the atmosphere, this method not only reduces the dependency on costly and environmentally harmful seeding materials but also aligns with the growing emphasis on sustainable and eco-friendly technologies in weather modification. In this regard, activating a substantial number of atmospheric aerosols into CCN represents a promising, low-cost, and environmentally friendly approach.

Water vapor clusters are easily formed around ions owing to the property of polar molecule, which has been confirmed through

extensive cloud laboratory experiments.[15–17] As early as 1899, Wilson found that X-ray irradiation can nucleate some negative ions to form small water droplets in the cloud chamber experiment when the supersaturation (SS) is 3 (i.e., relative humidity is 400%).[18] Leisner *et al.* investigated the interaction of laser-induced plasma channels with ice clouds and found that lasers can induce a multiplication effect of ice crystals in ice clouds.[19] Janhanshir explored the application of electric fields in weather modulation, particularly through techniques such as corona discharges to affect water droplets and ice crystals in clouds. It is shown that electric fields can change the charge state of water droplets, which in turn affects their condensation and freezing processes.[20] Ions and electrons produced by corona discharge can promote aerosol activation and increase the amount of CCN. Rabeony *et al.* studied the induced nucleation of vapors of different substances under the action of ions and electric fields in a cloud chamber and observed that both ions and electric fields could significantly promote nucleation.[21] As a supplement to exploring the effectiveness of ions as CCN in cloud chambers, the influence of ions generated by cosmic rays in the atmosphere on the formation of CCN has attracted the attention of scholars.[22–24] Harrison *et al.* studied ion-aerosol-cloud in the lower atmosphere, and the results showed that cosmic ray ionization is linked to cloud properties and there are electrical effects on aerosol and cloud microphysics.[25] However, the above-mentioned studies mainly focus on the effect of cosmic ray intensity on CCN. The potential of charged particles to enhance CCN concentration is a contentious issue, often attributed to the inherent unpredictability of atmospheric conditions. Despite numerous scholarly investigations into the employment of high-voltage-induced discharge for generating charged particles to catalyze precipitation, the observed phenomena have predominantly been condensation events rather than CCN formation.[26–29] It is plausible that such condensations are secondary outcomes of alterations in CCN concentrations. Presently, there exists a dearth of targeted research examining the impact of discharge on CCN levels and aerosol activation, a knowledge gap that, once addressed, could significantly advance the efficacy of warm cloud rainfall enhancement strategies.

Broadly speaking, charged particles are particles with an electric charge and have wide applications.[30–32] Currently, common ways to generate charged particles are femtosecond lasers, corona discharges, and plasma jets.[30,33] Femtosecond lasers are capable of releasing high energy in a very short period of time, but they are suitable for high-precision, small-scale experiments due to expensive equipment and high operating and maintenance costs. Corona discharge is less expensive, but the discharge is less stable and prone to ozone generation, which may cause secondary pollution.[1,10,34] Compared with the above-mentioned two methods, atmospheric pressure plasma jet (APPJ) excels in operation cost and stability,[35] and no harmful gases, such as ozone, are generated.[36–40] Therefore, this paper explores the effect of APPJ on the concentration of CCNs and aerosol activation through a cloud chamber.

II. EXPERIMENTAL SETUP

The experimental platform includes cloud chamber, charged particle generation system, and data measurement system, which are shown in Figs. 1(a) and 1(b).

The jet tube is placed at a height of 110 cm and is connected to an argon cylinder through a pipeline. Argon with a purity of 99.9% is selected as the working gas. The gas flow is controlled by a gas flow meter (MT-30A). A valve with a diameter of 2 cm is located at the bottom of the cloud chamber and is opened during argon injection to keep the pressure relatively stable. The copper electrode in the jet tube is connected to a high voltage pulse generator (DEI, PVX-4110) through a high voltage cable. The signal generator (Tektronix, AFG31102) and the DC power supply provide pulse signal parameters and voltage excitation for the high voltage pulse generator, respectively. The oscilloscope is connected to the current monitor and voltage monitor to record the voltage and current waveform. The temperature and humidity probe (Rotronic, HC2A-S) are placed near the top and bottom of cloud chamber to monitor temperature and humidity. Cloud condensation nuclei counter (CCNc, DMT CCN-100) is used to monitor the changes of CCN concentration. Aerosol spectrometer (Grimm 11-D) is adopted to measure the aerosols concentration. Sampling point A is at the same height with the jet pipe outlet. Sampling point B is below the point A. During the assessment of aerosol activation rates, the aerosol spectrometer samples the air from the cloud chamber, with its exhaust port connected directly to the air inlet pipe of the cloud condensation nuclei counter (CCNc). Conversely, when determining solely the concentration of CCN, the CCNc samples the air directly from the cloud chamber, bypassing the aerosol spectrometer housed within the green-framed enclosure.

The experimental ambient temperature is $21 \pm 0.2\,^{\circ}\mathrm{C}$, and the relative humidity is maintained at 85% ± 5%. We use DBD (dielectric barrier discharge) type jet plasma as the plasma source. The pulse voltage parameters are as follows: the peak voltage is 8 kV, the frequency is 8 kHz, and the pulse width is 2 μs. The discharge voltage–current curve is shown in Fig. 1(c). It can be seen that one voltage pulse has two current pulse signals, corresponding to the rising edge and falling edge of the voltage, respectively. The pulse current signal at the falling edge is caused by the charges accumulated outside the quartz tube during the first discharge at the rising edge.

III. RESULTS

A. Effect of plasma jet on the CCN concentration

The CCN counter (CCNc) was set to a supersaturation level of 0.4%. The supersaturation level here refers to the supersaturated operating environment of the CCNc's built-in cloud chamber created by the temperature gradient. To isolate the impact of the plasma jet, which is accompanied by argon gas injection, a control experiment was conducted with argon gas injection alone. This control involved maintaining an argon flow rate of 1.2 L/min. Subsequently, at sampling points A and B, the CCN concentration was measured across five distinct stages: static conditions, argon gas injection only, plasma jet activation, subsequent argon gas injection, and a return to static conditions. The experimental results are shown in Fig. 2.

Figure 2(a) shows the variation curve of CCN concentration in the cloud chamber at different times. Initially, at the static phase (0–15 min), the CCN concentrations at points A and B within the cloud chamber were comparable, suggesting that the cloud chamber's initial conditions were consistent across both experimental sets. Following the introduction of argon gas (15–30 min), a gradual decline in the CCN concentration was observed, likely due to gas dilution. Upon activation of the plasma jet (30–60 min), the CCN concentration initially

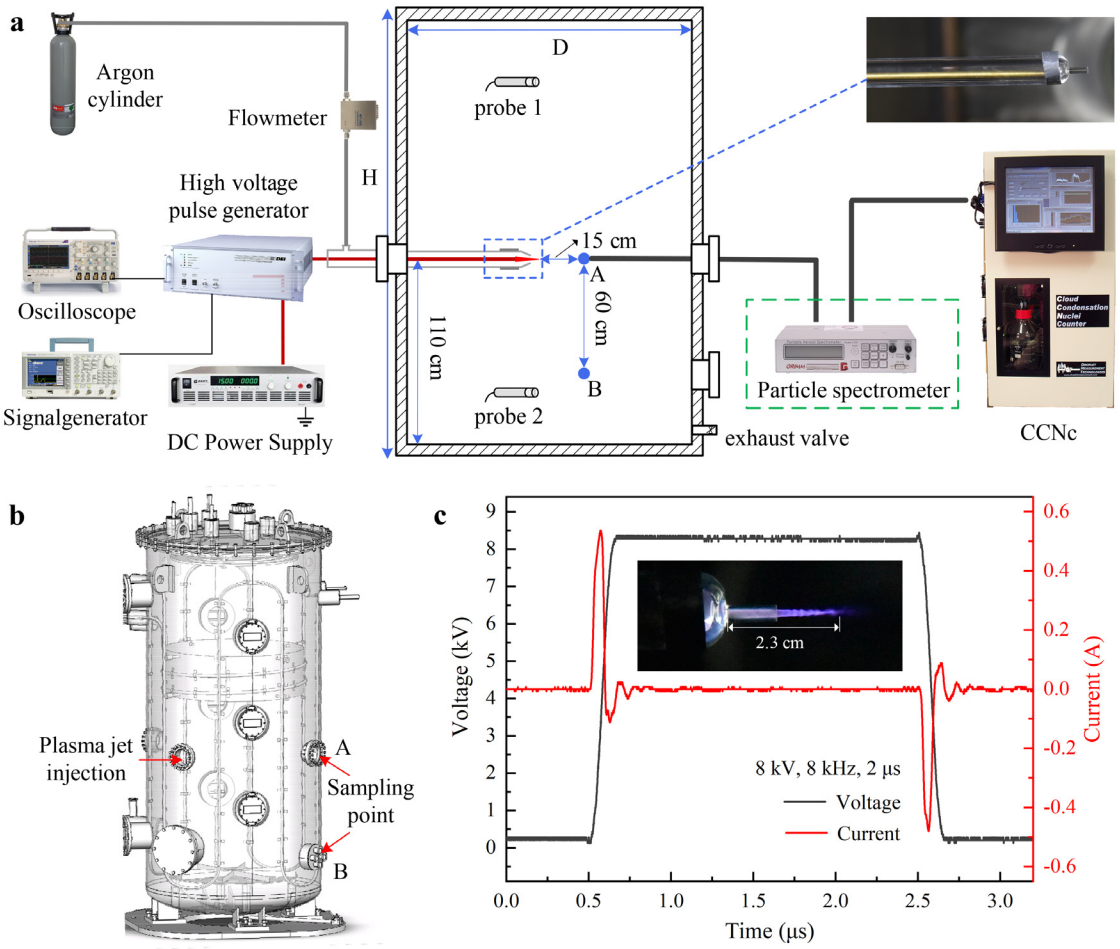

FIG. 1. (a) Schematic of the experimental chamber; (b) 3D diagram of the cylindrical cloud chamber; and (c) voltage–current characteristics of discharge. D = 1.0 m and H = 2.3 m.

persisted in decreasing, potentially influenced by the ongoing argon injection. However, a significant and rapid increase in the CCN concentration was noted after the plasma jet operated for an extended period. Specifically, after 30 minutes of plasma jet operation, the CCN concentration significantly exceeded the initial static levels, increasing by approximately 2.6-fold at point A and 1.1-fold at point B. Subsequently, when the plasma jet was deactivated and argon gas injection continued (60–75 min), the CCN concentration commenced to decline. Once the argon gas injection ceased, the CCN concentration stabilized. Notably, in comparison with the pre-discharge static state (0–15 min) and the post-discharge static state (75–90 min), the CCN concentrations at points A and B had risen by approximately 190% and 55%, respectively. The concordant experimental observations at points A and B confirm the plasma jet's efficacy in substantially elevating the CCN concentration within the cloud chamber.

Furthermore, this study examined the particle size distribution spectra of CCN at various experimental timestamps. At point A during sampling, with a supersaturation of $SS = 0.4\%$, the particle size distribution is depicted in Fig. 2(b). Observations revealed that the quantity of CCN across all particle sizes notably increased following the activation of the plasma jet, with a significant emergence of larger-sized CCN. This phenomenon suggests that the plasma discharge not only stimulates an increase in the quantity of CCN but also enhances their size. Larger CCN particles are crucial for facilitating the coalescence and growth of cloud droplets, which in turn can catalyze rainfall. These enlarged CCN particles compete with naturally occurring CCN and, due to their increased size and capacity to absorb water vapor, effectively diminish the cloud's supersaturation. This reduction in supersaturation leads to a decrease in the cloud droplet concentration, an expansion of the cloud droplet size spectrum, and ultimately promotes the collision and growth of larger cloud droplets into raindrops. Notably, the presence of exceptionally large CCN ($\sim 10\,\mu m$) can directly amplify the population of larger droplets at the tail end of the droplet size spectrum, hastening the broadening of the spectrum and enhancing the formation of substantial raindrops.

B. CCN activation rate

The CCN activation rate is used to characterize the adsorption of water molecules by aerosols. It is defined as the ratio of the total

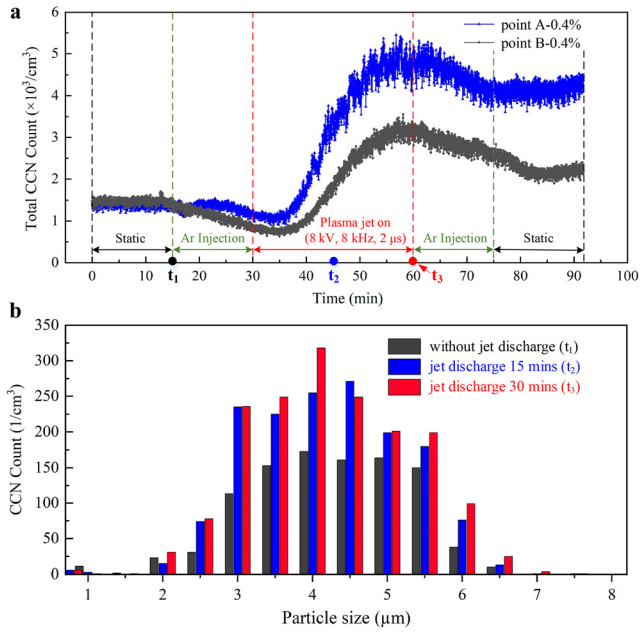

FIG. 2. (**a**) Supersaturation $SS = 0.4\%$, sampling at A and B. The number of CCN varies with time in the five stages of static; only argon gas injection; jet opening; only argon gas injection; and static; (**b**) particle size spectral distribution of cloud condensation nuclei at different times.

concentration of CCN to the total concentration of aerosols. Here, the total concentration of aerosols is defined as the total concentration of aerosols with particle sizes between 0.414 and 10 μm. The results of these measurements are depicted in Fig. 3, where the black line represents aerosol concentration, the blue line denotes CCN concentration, and the red line indicates the activation ratio. The data illustrate that upon plasma jet initiation, there is an immediate increase in the aerosol concentration. However, the CCN concentration lags, temporarily lowering the activation ratio. As the plasma jet operation proceeds, the activation ratio increases in tandem with the rising CCN concentration and subsequently declines once the jet operation ceases. Specifically, after 30 min of plasma jet operation, the activation ratio was observed to have increased approximately 2.5-fold. The aerosol concentration

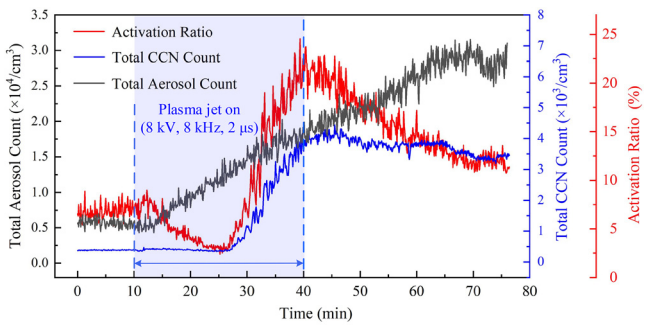

FIG. 3. The number concentration of CCN and aerosols, and the activation rate varied with time. Here, the activation rate is equal to the ration of total concentration of CCN and the total concentration of aerosols.

continued to ascend, reaching a plateau 25 min post-jet deactivation. This behavior is attributed to the fact that only aerosols larger than 0.414 μm are considered in the concentration measurements. Smaller aerosols below this threshold can merge with electrons or ions, thereby enhancing the growth of aerosols above 0.414 μm and leading to a sustained increase in the aerosol concentration. Moreover, the plasma jet introduces a substantial number of ions and electrons into the atmosphere, some of which aggregate and grow into larger aerosols. This process is a significant mechanism for the genesis of new atmospheric aerosol particles.[41,42]

IV. DISCUSSION

To elucidate the influence of plasma jets on aerosols and CCN, we measured the emission spectrum of the argon plasma jet using a high-sensitivity micro-spectrometer (SR-500I-A) with an optical resolution of 0.03 nm. The spectrometer's optical fiber probe was positioned 6 mm along the longitudinal axis and 10 mm laterally from the central axis of the jet tube outlet. The resulting spectrogram is shown in Fig. 4. The argon plasma jet emits a substantial quantity of electrons, hydroxyl (OH) radicals, argon ions (Ar^+), and other ion species. These emitted electrons, ions, and reactive radicals exert a beneficial influence on aerosol formation and activation.[9] In Fig. 5, we demonstrate four reaction pathways for CCN formation: ion mediated nucleation

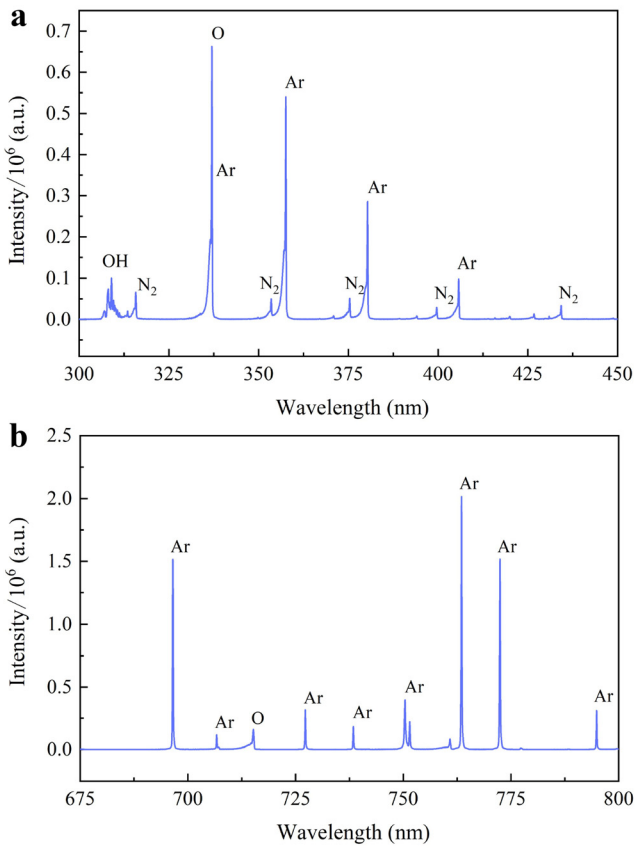

FIG. 4. Emission spectrum of argon (Ar) plasma jet.

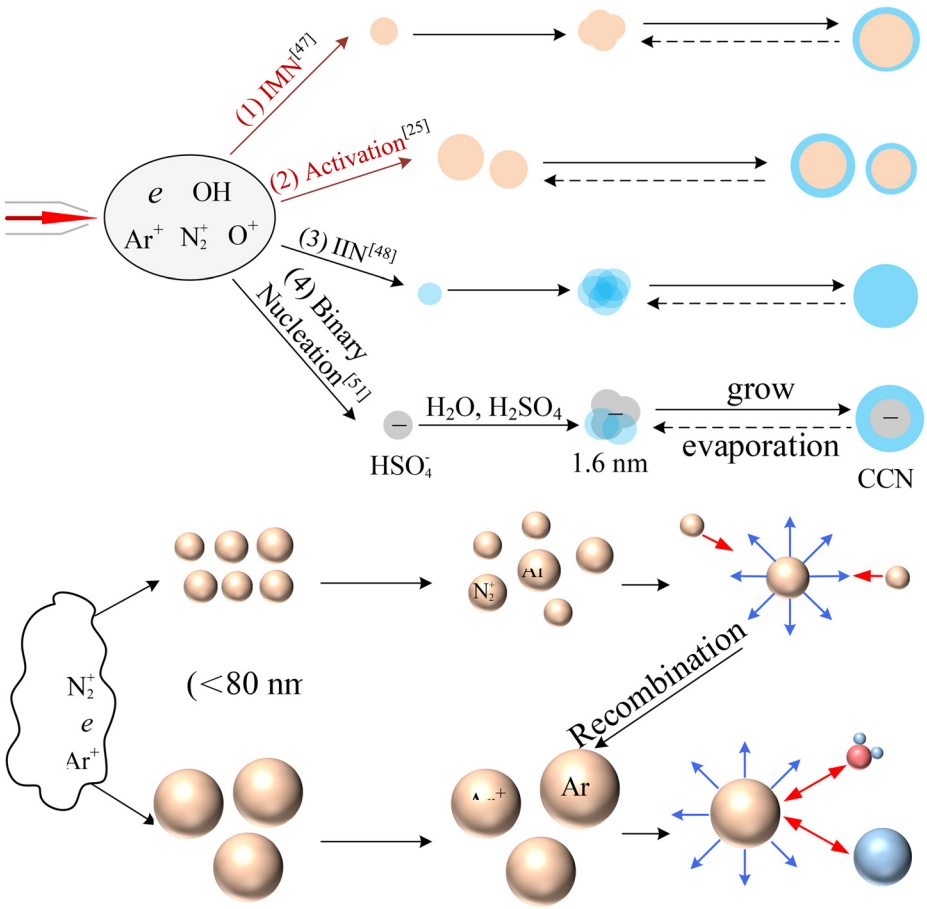

FIG. 5. (a) Schematic diagram of the effect of the charged particles generated by the plasma jet on the number of condensation nuclei and (b) the more detailed reaction pathway in (1) and (2), on the interaction of charged particles with ultrafine and large aerosols.

(IMN), activation, ion-induced nucleation (IIN), and binary nucleation. The detailed mechanisms are described as follows:

(1) **Ion-mediated nucleation (IMN)**. Field-induced and diffused charge pairs promote the growth of ultrafine aerosols. A substantial population of ultrafine aerosol particles, measuring less than 80 nm in diameter, is suspended within the cloud chamber. Upon activation of the plasma jet, these particles undergo charging processes, which include field-induced and diffusive charging mechanisms. Empirical evidence from various studies has established that charged particles experience an increased propensity for collision.[43–45] Such collisions among ultrafine aerosols are conducive to ion-ion recombination, thereby leading to the formation of aerosols with larger diameters, sufficient to reach CCN size thresholds, typically exceeding 80 nm. While the direct observation of nanoparticles remains challenging within experimental confines, the aerosol concentration, as depicted in Fig. 3, is noted to ascend during the plasma jet discharge phase, specifically between 10 and 40 min. This escalation in concentration can be attributed to the transformation of aerosols initially smaller than 0.414 μm into larger sizes that surpass this threshold, suggesting an enlargement of aerosol particles subsequent to the plasma discharge. In addition, scholars generally characterized the ease with which the aerosol size attracts water vapor to form CCN by the droplet saturation ratio.[12] By correcting the Kelvin equation, the previous authors found that the saturation ratio is inversely proportional to the particle size when the particle size of the charged droplets exceeds 1 nm.[12] Therefore, the increase in the aerosol size is conducive to attracting water vapor to condense and forming CCN.[46] This process of forming CCN by promoting already existing ultrafine aerosols is called ion-mediated nucleation.[47]

(2) **Activation.** Diffused charges lower the free energy barrier for the condensation of water vapor. Some of the aerosols within the cloud chamber naturally exceed 80 nm in size. Predominantly, these larger aerosols become charged through diffusion and subsequently collide with electrons or ions. This "charge effect" will reduce the free energy barrier in water vapor condensation, and the free energy barrier decreases with the carried charge. Furthermore, since water is a polar molecule, it creates an image charge when it is close to charged particles, which causes it to condense more easily on aerosols under the action of electrostatic forces.[44] Accordingly, within an identical supersaturated environment, similarly sized aerosols are more prone to attracting water vapor for condensation and activation as CCN once charged.[25] This observation aligns with the

notable escalation in the activation ratio of aerosols to CCN following plasma jet discharge, as depicted in Fig. 2(b).

(3) **Ion-induced nucleation (IIN)**. Ion-induced nucleation promotes the formation of cloud condensation nuclei. Water vapor typically condenses to form droplets at exceedingly high supersaturation levels. However, the presence of ions can significantly enhance the condensation process, a phenomenon known as ion-induced nucleation.[12,48] Ions lower the surface free energy required for water vapor condensation,[49] thereby facilitating the formation of charged droplets on ionic surfaces. These charged droplets exhibit an increased affinity for neutral water vapor, elevating the likelihood of collision with additional water vapor molecules.[50] This enhancement in collisions not only promotes nucleation but also encourages the aggregation with surrounding water vapor molecules, ultimately leading to the growth of these droplets into CCN.

(4) **Binary nucleation.** The plasma jet produces a high concentration of hydroxyl radicals, which promotes the formation of new aerosols. Throughout the plasma jet discharge, a heightened concentration of hydroxyl (OH) radicals is produced. These OH radicals possess unpaired electrons in their outermost electron shell, endowing them with a high reactivity and a propensity to seek additional electrons. Consequently, they serve as potent oxidizing agents capable of transforming trace atmospheric components, including the oxidation of sulfur oxides. This oxidation sequence culminates in the formation of acidic species, such as sulfuric acid and its salts, along with aerosols. This sequence of events is categorized as binary nucleation.[51] Given that the cloud chamber's air composition mirrors that of the ambient environment, it invariably contains trace levels of sulfur dioxide (SO_2). Under the influence of OH radicals, SO_2 is converted into sulfate particles, which may further evolve in accordance with the processes outlined in reaction pathway (1). This recurring atmospheric mechanism is pivotal for the genesis of new aerosol particles.

In this study, we observe a rapid and sustained increase in the population of aerosols larger than 0.414 μm following the plasma discharge, primarily attributed to the collision and subsequent growth of aerosols initially smaller than 0.414 μm. Concurrently, the activation rate of aerosols exhibits a swift upsurge upon the initiation of plasma jet activity. Thus, the data suggest that the observed augmentation in CCN counts is predominantly driven by reaction pathways (1) and (2), as illustrated in Fig. 5.

The experimental data consistently demonstrate that the concentration of CCN within the cloud chamber remains elevated even after the plasma jet operation is terminated. This phenomenon can be attributed to the charged aerosols' ability to diminish the level of supersaturation necessary for water vapor condensation. Specifically, the partial pressure of water vapor necessary to sustain the liquid phase of droplets is diminished, consequently decreasing the evaporation rate of water vapor from the CCN. This observation aligns with the equilibrium partial pressure of water vapor following droplet charging as reported by Rusanov *et al.*[52] Furthermore, the presence of ions in the chamber aids in the stabilization of molecular clusters, thereby enabling the prolonged maintenance of a high CCN concentration within the cloud chamber.

In the future application for real clouds, aircraft (unmanned aerial vehicle, airship, etc.) can be equipped with APPJ array under suitable conditions. Vehicles with APPJ arrays are driven into warm clouds, and while generating plasma jets, appropriate amounts of hygroscopic salts, aerosols, and other substances are uniformly mixed into the jets, which are emitted into the warm clouds. These substances can act as condensation nuclei to promote the condensation of water vapor to form larger water droplets, which in turn initiate or accelerate the precipitation process in the warm clouds.

V. CONCLUSION

Within the scope of this manuscript, we have constructed a cloud chamber platform to meticulously examine the impact of plasma jet discharge on both the concentration of CCN and the activation rate of aerosols. The key findings and subsequent analysis from our experimental series are concisely encapsulated as follows:

(1) Plasma jet discharge significantly elevates the concentration of CCN. Upon maintaining the plasma jet operation for 30 min, we observed a substantial increase in the CCN concentration, ranging from 1.1 to 2.6 times higher than that of the undischarged static state.
(2) Following the termination of plasma jet operation, the CCN concentration within the cloud chamber is sustainably maintained at an elevated level. A comparison with the predischarge static stages reveals an enhancement in the CCN concentration by 55%–190%.
(3) Plasma jet discharge notably augments the transformation of aerosols into CCN. The activation rate soared to approximately 2.5 times the original level after the plasma jet was activated for a duration of 30 min.

The experimental outcomes affirm the viability of employing plasma jet technology to enhance CCN formation and aerosol activation. This method exhibits substantial practical utility and holds profound implications for the realm of artificial weather modification.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Dingchen Li and Ming Zhang contributed equally to this paper.

Chuan Li: Conceptualization (lead); Writing – original draft (lead); Writing – review & editing (lead). **Tingyu Liang:** Conceptualization (equal). **Zutao Wang:** Data curation (equal). **Pengyu Wang:** Data curation (equal). **Ming Zhang:** Conceptualization (equal). **Yong Yang:** Conceptualization (equal). **Kexun Yu:** Conceptualization (equal). **Yuan Pan:** Conceptualization (equal). **Dingchen Li:**

Conceptualization (lead); Writing – original draft (lead); Writing – review & editing (lead).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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