Separating Cloud Forming Nuclei from Interstitial Aerosol

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1. Introduction

Our poor representation of aerosol and cloud interactions in the climate models have led to the largest uncertainty in predicting climate change. Studies have shown that CN can influence climate by changing the properties of clouds. Aerosol particles that act as CN can be broadly classified based on their source into two categories: natural and anthropogenic aerosol. The global source strength of natural aerosol is higher than anthropogenic aerosol; however, certain specific anthropogenic constituents can amplify the aerosol effect on clouds. In addition, the atmospheric trace amounts of soluble gases and organic substances can alter the aerosol properties from both of the sources.

Recent studies have shown that various aerosol properties (size, surface chemistry [hygroscopicity and wettability] and active sites) as a function of temperature and humidity can determine the CN efficiency of aerosol. Atmospheric scientists are working towards finding a relationship between these properties to parameterize the observations in the climate models. But without an adequate knowledge of CN properties this representation cannot be improved further.

The technique of CN separation from the interstitial aerosol has the advantage that by measuring the specific properties of CN, simplifies the model representation task. For example, laboratory and *in-situ* techniques can be used to differentiate the CN in CCN and/or IN measurements and their properties can be measured. Therefore, the modelers can narrow down the physicochemical properties of CN to be incorporated into the representation task. Further, the information of the aerosol chemistry helps to determine aerosol source: natural versus anthropogenic.

2. Separation techniques

The separation of CN from interstitial aerosol technique is based on the particle's inertia. The instrument that employs this technique is called counterflow virtual impactor (CVI). The separation is achieved by stopping and removing the gas phase and small particles but capturing large particles with sufficient inertia to cross gas streamlines. Particles with insufficient inertia to be captured follow the deflected streamlines and are removed from the system. Higher inertia particles are injected into a typically clean, dry, and warm counterflow carrier gas that causes evaporation of condensed phase water. This technique has the advantage that a broad cut size range can be achieved by varying the flow rates associated with the CVI without changing the physical dimensions of the instrument.

The CVI used in the laboratory set up is called pumped CVI (PCVI) and the CVIs used for *in-situ* measurements are called airborne CVI (ACVI). The flow schematics of these designs are shown in Figure 1 a) and b), respectively. In PCVI design the aerosol particles are pulled inside the instrument and undergo inertial separation, while in ACVI the aircraft velocity imparts motion for aerosol particles that are again separated based on the inertia. Both designs are widely used and their performance characteristics are documented.

3. Design considerations

The CVI's performance is characterized by a particle collection efficiency curve. In an ideal environment, the separation between the CN and interstitial particles should be perfectly sharp. However, due to the non-idealistic flow behavior within the CVI, the true efficiency is hardly achieved.

Figure 2 shows the particle transmission efficiency (TE) of ammonium sulfate particles as a function of its size [1]. Three different flow configurations were used, implying the importance of relationship between the flows. It can be observed that by varying the flows, different sizes of particles can be sampled, but, as mentioned above, due to non-idealistic flow behavior, the TE do not reach 100%: imperfect TE and also sharp TE are not observed. It was suggested that imperfect TE is caused because the particles near the wall surface are trapped in the recirculation zone and do not join the sample flow. Also, because the flow within the counterflow region is not well-developed. This flow heterogeneity allows the small particles to penetrate the counterflow region and join the sample flow, even though they should be rejected.

The particle TE can be theoretically calculated based on the following equation [2],

$$L = K.r.\frac{\rho_p}{\rho_g}.\left(Re^{1/3}.C^{1/2} - \frac{\pi}{2} + \varphi\right) \tag{1}$$

Where,

$\varphi = tan^{-1}\left(Re^{-1/3}.C^{-1/2}\right); 0 \leq \varphi \leq \frac{\pi}{2},$ L is the stopping distance, K is a constant (= 5.3075), r is the radius of the particle, ρ_p is the density of the particle, ρ_g is the density of the flow media, Re is the Reynolds number, and C is a constant (= 0.158). For the desired flow configuration, if the distance between the tip of the CVI nozzle till the beginning of the sample flow is larger than the particle stopping distance, then the particle joins the sample flow and is transmitted. Figure 3 shows the relationship between the droplet diameters to the stopping distance for different flows. The flow can be varied either by increasing the input flow, while keeping other flows constant, or by varying the CVI geometry. The former option is always desired.

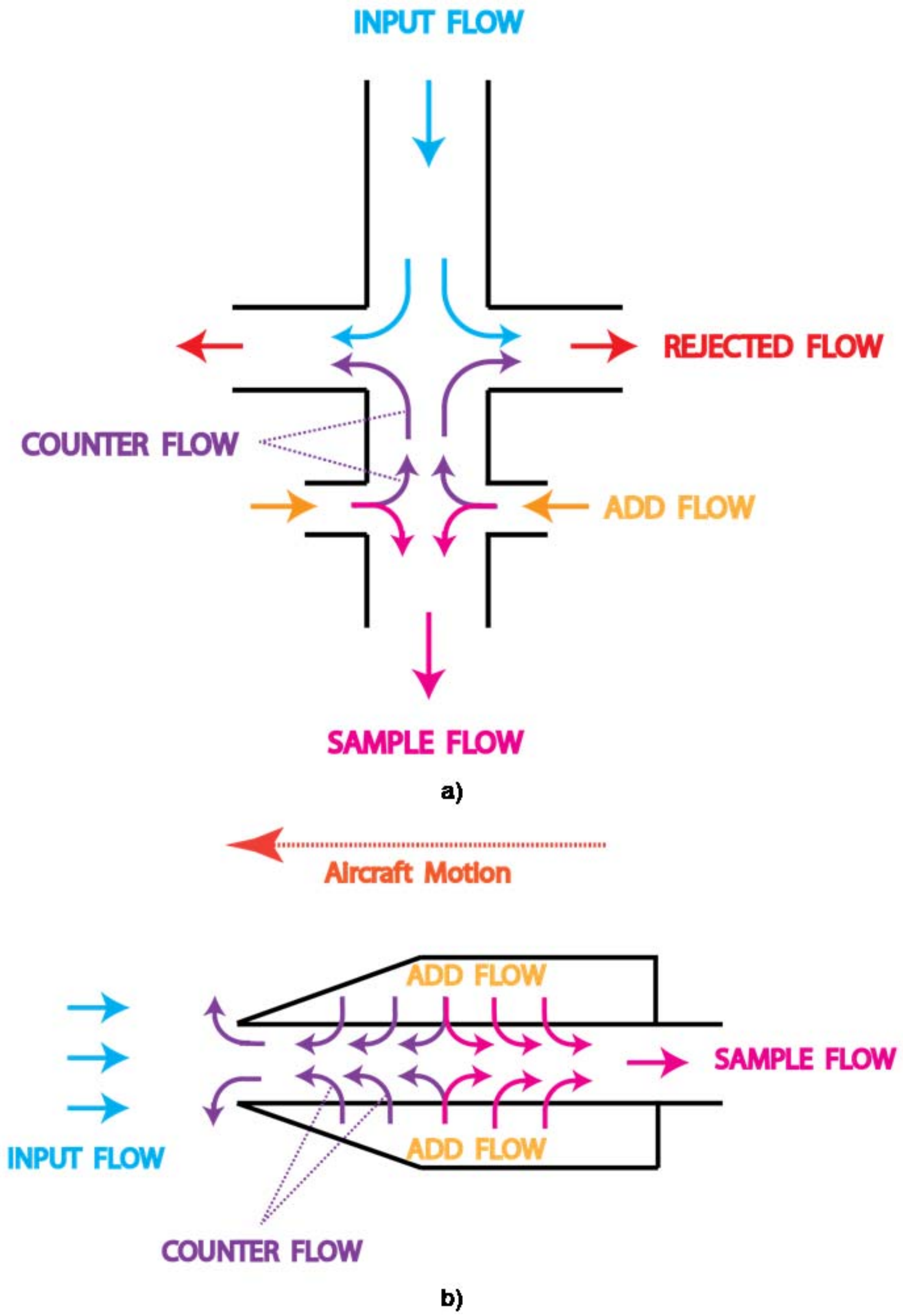

Figure 1. a: Flow schematics within the pumped counterflow virtual impactor (PCVI). Input flow carries the condensation nuclei and interstitial particles (non-activated aerosol particles). The large particles that have sufficient inertia to cross the streamlines enter the counterflow; these particles that

can overcome the counterflow, join the sample flow. The remaining particles join the rejected flow. The sampled condensation nuclei (CN) are then forwarded to the respective analytical tools. The add flow consists of dry and particulate-free gas that splits into counterflow and sample flow. The counterflow then joins the input flow to become the rejected flow. In normal PCVI operation, the sample flow is maintained constant while the counterflow is varied to sample various CN sizes. b: Flow schematics within the airborne counterflow virtual impactor (ACVI). The motion to the CN and interstitial particles is given by the motion of the aircraft. This induces the input flow and therefore, indirectly, the inertia to the particles. Similar to the PCVI operation, the counterflow rejects the particles that do not have sufficient inertia to overcome the counterflow; the particles having sufficient inertia join the sample flow. The sample particles are then forwarded to the desired analysis tools. Again, similar to the PCVI, the add flow splits into counterflow and sample flow, and thus, by varying the add flow, various sizes of CN can be sampled (assuming sample flow is maintained constant).

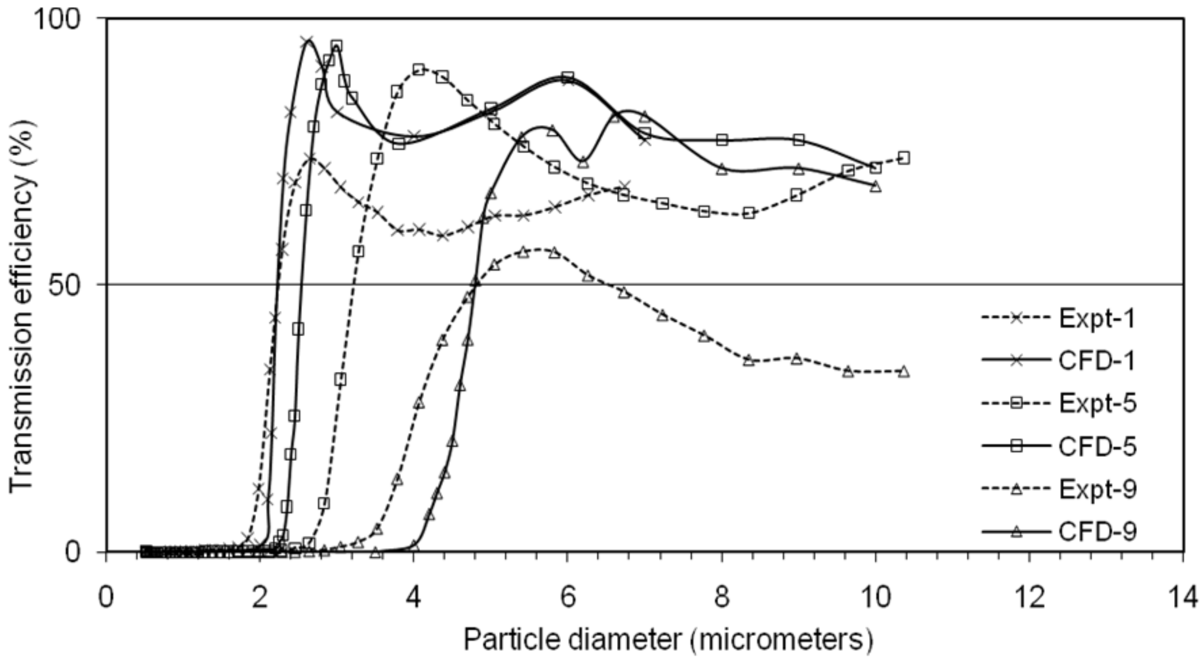

Figure 2. Particle transmission efficiency (TE) of ammonium sulfate particles. Experimental (Expt) TE are compared to computational fluid dynamics (CFD) predicted TE for various cases where PCVI flow configuration was varied [1].

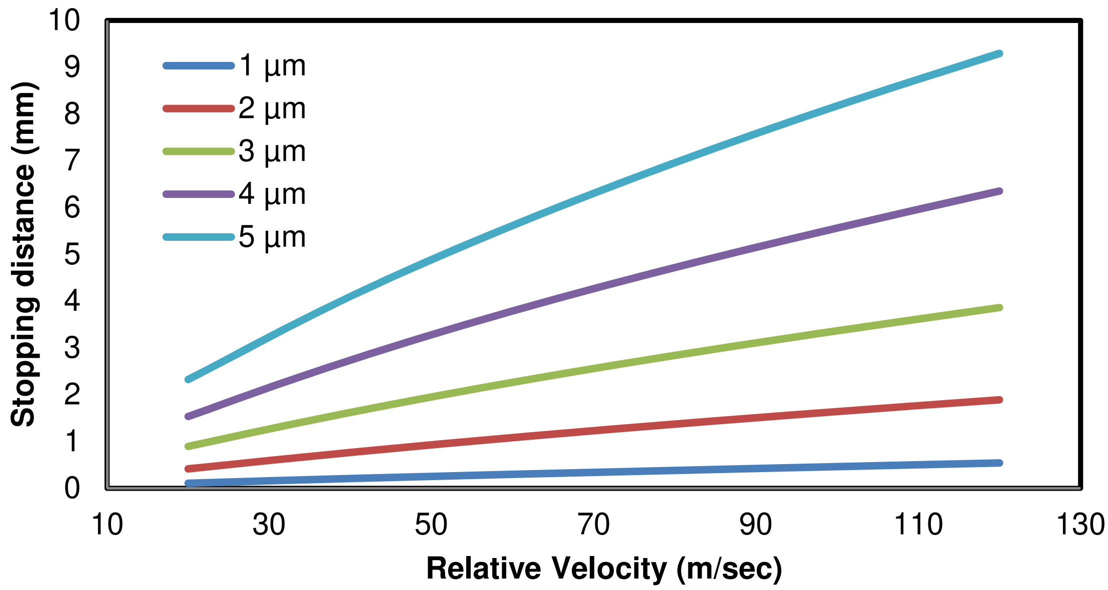

Figure 3. Stopping distance of various-size water droplets as a function of their relative velocity with respect to flow media when exiting the nozzle (in PCVI) or entering the counterflow region (in ACVI). If the physical dimensions of the CVI are known, then these relationships can be used to determine the theoretical TE of the particles.

4. Operational challenges

As discussed above, the PCVI performance characteristics depend upon flow behavior and geometrical design. Recently, limitations and uncertainties associated with the CVI's have been identified [1]. They include particle losses at walls, imperfect transmission efficiencies of CN, limited size range of transmitted particles, turbulence effect on the droplet breakup and shattering, and narrow range of measurement flow rates. To understand the artifacts and improve further the designs, CFD simulations were carried out. For example, fluid flow characteristics of PCVI are analyzed to understand the performance characteristics and associated artifacts, as shown in Figure 4.

The white colored particle deposition on the walls can be observed (Fig. 4 top panel). The particle deposition losses occur at various eddies and vortices shown in the bottom panel of Fig. 4. The magnified CFD image shows eddies and recirculation vortices generated as a result of the flow boundary conditions and the PCVI design geometry. Such CFD simulations are necessary to improve the design to reduce the particle losses.

Under the influence of flow turbulence within the CVI instrument, it is possible that when sampling cloud, hydrometeors (liquid droplet and ice crystals) can break or shatter leading to numerous small particles. This is undesirable as the particle TE will be reduced and might lead to non-conclusive results. It has also been observed that at high airspeeds (in airborne CVI), large drops and ice crystals can impact on the probe inlet and break; this also happens within the counterflow region (because of shock). Droplet breakup criterion is usually calculated using Weber number: $We = (V_g - V_{drop})^2 \cdot \rho_g . d_{drop} / \sigma$, where V is the velocity of the gas

(g) and droplet (d; drop). The droplet breaks when the We number is greater than 12. These estimates can be validated by combining the observations and CFD simulations.

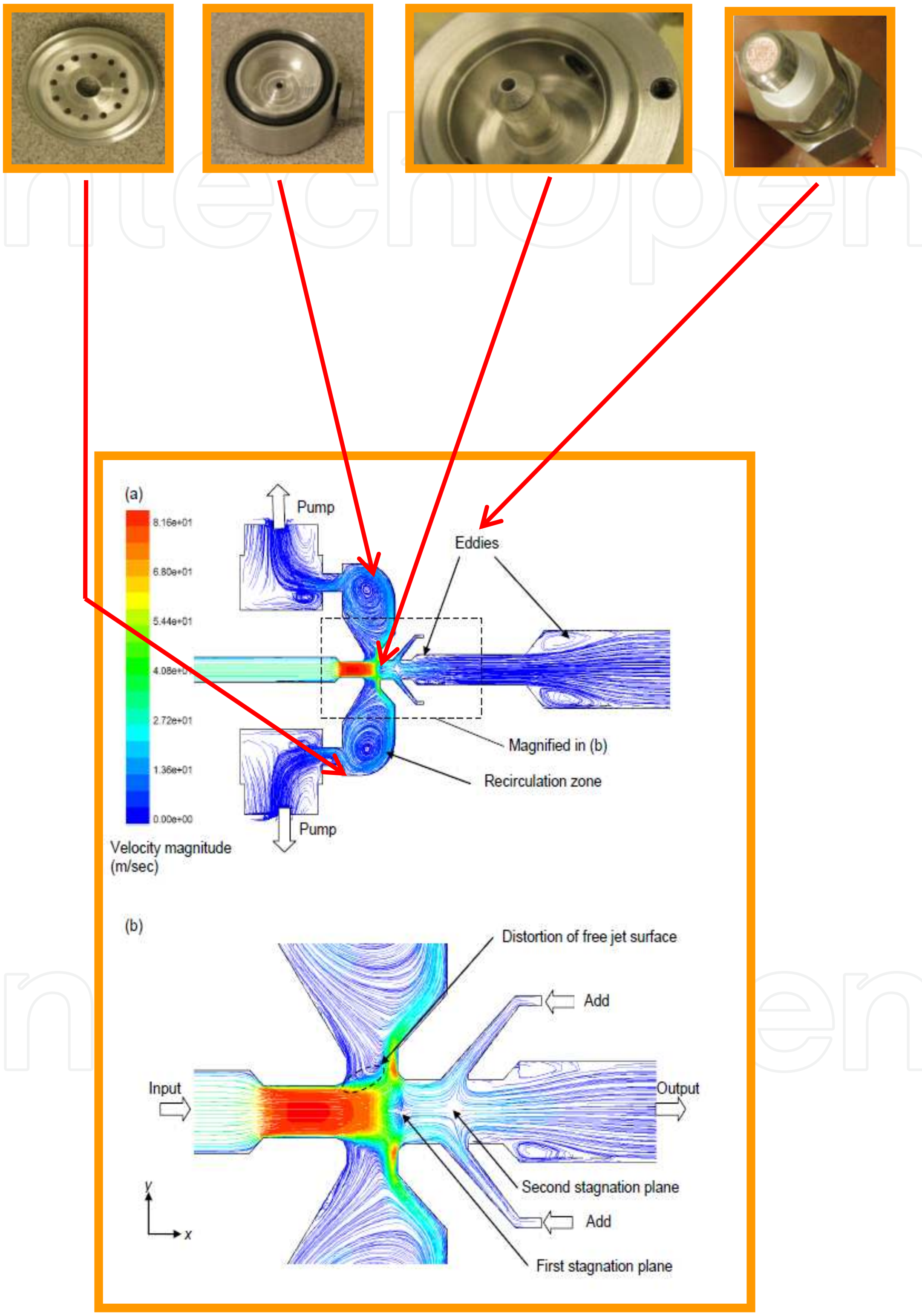

Figure 4. Interior surfaces of the PCVI instrument showing (upper panel) the particle deposition regions. These regions are indicated within CFD predicted velocity pathlines (bottom panel). See text for details [3].

Another feature that might be important, which is not well-documented, is the scavenging of the interstitial aerosols when sampling cloud droplets. Due to the mass differences between these two types of particles, the relative velocity could become significant and could lead to collision between the droplet and interstitial aerosol. If the droplet collides with the interstitial particles, then these particles might get trapped within the droplet and if this droplet gets transmitted, that will yield undesired results. As of now, non-activated aerosol particles (interstitial aerosol) are being characterized as activated aerosol particles, but this is not correct. Systematic studies are required where water droplets and interstitial aerosols should be generated and their collision efficiency should be investigated.

5. Summary

In this chapter, a technique that separates the cloud forming nuclei from the interstitial aerosols is briefly discussed. The technique is based on the inertia of the particle. Cloud forming nuclei are the residual particles of the droplets and ice crystals. These cloud hydrometeors have high inertia compared to the interstitial aerosols and therefore penetrate the counterflow region of the CVI to be sampled. Two types of CVI instruments are based on this technique: PCVI and ACVI. PCVI is generally used in the laboratory set-up where the particle velocity is achieved by pumping the input flow; whereas, in the ACVI, the particle velocity is generated by aircraft flight.

Transmission efficiency of the particles that are sampled can be theoretically calculated, and it was observed that as particle velocity and/or its diameter increases the efficiency also increases. Several artifacts of the cloud separation technique are described. They include particle losses and imperfect transmission efficiencies, flow turbulence effects on the droplet breakup and shattering, and possibility of scavenging of interstitial aerosols (this needs further investigation). However, many studies have quantified these artifacts and the cloud separation technique is now considered as a must have measurement platform for most of the laboratory and field studies.

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6. References

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