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# CCN Closure and Composition Analysis of Droplet-Forming Aerosol

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**Abstract.** Cloud condensation nuclei, aerosol chemical composition, and aerosol size measurements were determined at a field site subject to a variety of aerosol sources. A pumped counterflow virtual impactor was utilized to directly determine the chemical composition of the droplet activating aerosol.

**Keywords:** Cloud Condensation Nuclei, clouds, aerosols
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## INTRODUCTION

Interactions between aerosol particles and clouds contribute significantly to Earth's radiative budget [1, 2]. Aerosol particles can act as cloud condensation nuclei (CCN) wherein particles are able to activate by the accumulation of water until they grow to become cloud droplets, which can modify the microphysical properties of warm clouds and number concentration of cloud droplets, impacting indirect effects on climate [3, 4]. Current assessment of interactions between aerosols and clouds is uncertain and parameters used to estimate cloud droplet formation in global climate models are not well constrained [1]. In order to quantify, constrain, and predict the radiative effects of warm clouds, it is crucial to understand the connections between the chemical composition of the preexisting aerosol particles and their propensity to form cloud droplets under various meteorological conditions.

Whether an aerosol particle acts as a CCN at a given atmospheric supersaturation is mainly determined by its size, or dry diameter, and its ability to take up water before activation into a cloud droplet, or hygroscopicity [5, 6]. Factors determining CCN activation include the particle size, mixing state (internal vs. external), chemical composition, amount of soluble material in the particle, and atmospheric saturation conditions [7, 8].

Ambient aerosol particles are composed of complex mixtures of inorganic and organic components, leading to difficulty in determining their hygroscopicity and CCN activation. However, while many laboratory studies have succeeded in characterizing the CCN activation of single component aerosol particles [9], as well as

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particles with simplified mixing states [10, 11, 12], translating these laboratory studies to the real atmosphere has proved challenging. While the aerosol number size distribution is crucial in characterizing CCN activity [7], particle composition needs to also be considered, especially in considering the effects of atmospheric processing on aerosol particles in transport, such as oxidation, condensation of trace gases, and heterogeneous chemistry [13, 14, 15]. A recently developed hygroscopicity parameter, kappa, combines aerosol chemistry and size information to represent the hygroscopic behavior of multi-component aerosol particles, which are more typical of complex aerosol particles found in the atmosphere [9].

Kappa values from laboratory experiments using simplified aerosol mixtures of atmospheric relevance range from 0.5 to 1.4 for highly soluble salts (e.g., ammonium sulfate, sodium chloride, which are commonly observed in ambient particles), and 0.01 to 0.5 for a range of organic species [9]. The challenge exists in extending laboratory measurements of CCN to atmospheric conditions, thus field studies which allow for ambient measurements in a variety of conditions and environments impacted by a variety of different particle sources (e.g. urban emissions, terrestrial biogenic emissions, marine emissions) are important in assessing our understanding of atmospheric CCN and associated activation properties [16, 17, 18, 19].

The important parameters that contribute to CCN activity, such as size, composition, mixing state, and hygroscopicity, can be utilized to predict CCN concentrations at given atmospheric conditions, which are then compared to direct observations of CCN number concentrations in what has become known as a CCN "closure study". In this way we can assess our understanding of the interactions between hygroscopicity, size, chemical composition, and CCN activation [20]. We present a CCN closure study as well as direct composition analysis of CCN activating aerosol CCN measurements at a field site subject to a variety of airmass sources.

## METHODS

Measurements were conducted February 2013 as part of the DOE Two Column Aerosol Project (TCAP). Located in North Truro, MA, the site provides access to a variety of air mass sources, including marine, aged continental, and aged urban aerosol.

A CCN closure study was conducted with measurements from a commercial Cloud Condensation Nuclei Counter (CCNC, Droplet Measurement Technologies) at a range of supersaturations, as well as an Aerosol Mass Spectrometer (AMS, Aerodyne). Further measurements were conducted utilizing a Pumped Counterflow Virtual Impactor (PCVI) in order to separate the activated droplets from un-activated aerosol at the output of the CCNC, with subsequent composition analysis of the residual droplet aerosol by the AMS.

The PCVI uses a vacuum pump to draw particles into the inertial separation unit; a particle-free flow (counterflow) opposes the particle flow. The counterflow acts as a barrier against particles below a prescribed cut-size; these particles are removed by the pump flow and particles greater than the cut-size are able to overcome the counterflow and become entrained into the sample flow. The rate of counterflow can be adjusted to vary the cut-size of particles entering the sample flow, and thus can be set so as only

cloud droplets are able to overcome the counterflow while submicron unactivated particles are removed by the pump flow. The particle-free dry nitrogen gas dries the droplets at the exit of the PCVI, allowing the particles that seeded these cloud droplets to be sent to other instrumentation for analysis [21, 22, 23]. A schematic is shown below.

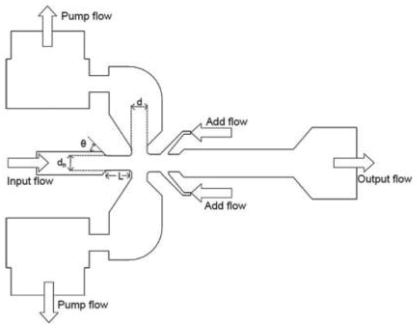


**FIGURE 1.** schematic diagram of the PCVI geometry showing the different flows [23]. The add flow corresponds to the counterflow, which sets the cutpoint size/inertial barrier for particles. Particles smaller that cannot overcome the inertial barrier are removed by the pump flow. The output flow containing particles large enough to overcome the inertial barrier is then sent to the AMS for composition analysis.

The PCVI has been previously applied at the output of a commercial CCNC in the laboratory [22], and in the field at the output of a custom-built CCN chamber [24]. Both studies involved an AMS to determine the chemical composition of the residual activated droplets. Supersaturations studied with this system ranged from 0.3% to 0.7%, and results will be presented also as a function of airmass source and comparison with the total ambient aerosol population.

## RESULTS AND DISCUSSION

Presented results include CCN concentrations and the associated activated fraction of the total aerosol population into droplets. Concurrent AMS composition and CCN concentration measurements were utilized in a CCN closure study to predict CCN concentrations as a function of airmass source and CCN chamber supersaturation, and assess the degree of chemical composition knowledge necessary to achieve closure. Back trajectories and aerosol composition were also utilized to assess aerosol age and extent of processing.

High-resolution composition measurements of droplet residual particles were determined at a range of supersaturation values and compared to the total ambient aerosol composition. Constraints and advantages of the CCNC-PCVI-AMS system also were assessed.

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