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DOE/SC-ARM-TR-225

Cloud Condensation Nuclei Particle Counter Instrument Handbook – Airborne Version

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September 2019

Work supported by the U.S. Department of Energy, Office of Science, Office of Biological and Environmental Research

Acronyms and Abbreviations

ARMAtmospheric Radiation MeasurementCCNcloud condensation nuclei counterDOEU.S. Department of EnergyNISTNational Institute of Standards and TechnologyOPCantical secticle security	ACE-ENA	Aerosol and Cloud Experiments in the Eastern North Atlantic
DOEU.S. Department of EnergyNISTNational Institute of Standards and Technology	ARM	Atmospheric Radiation Measurement
NIST National Institute of Standards and Technology	CCN	cloud condensation nuclei counter
	DOE	U.S. Department of Energy
ODC service la service la service de la serv	NIST	National Institute of Standards and Technology
OPC optical particle counter	OPC	optical particle counter
OSS Operation Status System	OSS	Operation Status System
PC personal computer	PC	personal computer
SMPS scanning mobility particle sizer spectrometer	SMPS	scanning mobility particle sizer spectrometer
SS supersaturation	SS	supersaturation
USB Universal Serial Bus	USB	Universal Serial Bus
UTC Coordinated Universal Time	UTC	Coordinated Universal Time
VGA video graphics array	VGA	video graphics array

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1.0 Instrument Title

Model CCN-100 or CCN-200 cloud condensation nuclei counter

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4.0 Instrument Description

The cloud condensation nuclei counter (CCN; Figure 1) is a U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) user facility instrument for measuring the concentration of aerosol particles that can act as cloud condensation nuclei (1, 2).

Note: ARM deploys both ground and airborne versions of this instrument. This handbook covers only the airborne instrument; the ground version is covered by <u>DOE/SC-ARM-TR-168</u>. The main differences

between the two are that the airborne CCN operates at fixed supersaturation and adapts thermal efficiency calculation in the data process.

The CCN draws sample aerosol through a column with thermodynamically unstable supersaturated water vapor that can condense onto aerosol particles. Particles that are activated, i.e., grown larger in this process, are counted (and sized) by an optical particle counter (OPC). Thus, activated ambient aerosol particle number concentration as a function of supersaturation is measured. Models CCN-100 and CCN- 200 differ only in the number of humidifier columns and related subsystems: CCN-100 has one column and CCN-200 has two columns along with dual flow systems and electronics.



Figure 1. The cloud condensation nuclei counter (CCN-100 with an optional computer display). Image adapted from the manufacturer's website.

5.0 Measurements Taken

The main measurement outputs of the CCN are the total number, concentration, and size of humidified (activated) aerosol particles (droplets) as a function of supersaturation in the humidifier. Additional measurements include sample flow rate, sample air pressure, and sample temperature.

6.0 Links to Definitions and Relevant

6.1 Data Object Description

The data from the CCN are recorded in plain text in column format with appropriate headers. The data fields recorded include:

Measurement date/time, supersaturation, humidifier temperatures, system temperatures, sample flow, sheath flow, sample pressure, laser current, OPC monitor voltages, particle number concentration by size bin (20 bins), and total particle number concentration.

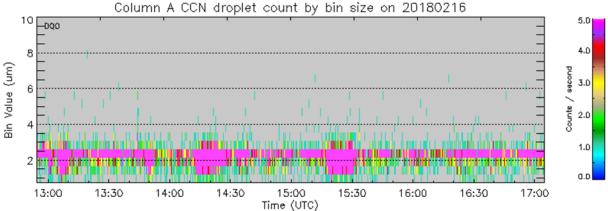
Data are recorded after every sample, typically every second. A new data file is started every hour and every time the system is restarted.

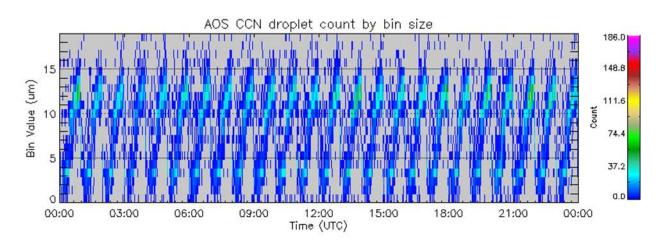
Data Ordering 6.2

Data from the CCN can be ordered from http://www.arm.gov/instruments/ccn-air. Data are organized by measurement location/campaign.

Data Plots 6.3

Figures 1 and 2 show typical data for the CCN. Figure 1 shows measured size distributions (particle counts per size bin) of activated aerosol particles as a function of time. The periodic nature of the plot is due to the humidifier supersaturation being periodically changed, which affects the final size of the activated particles (droplets). Figure 2 shows total particle number concentration as a function of time with data points colored by current supersaturation % (SS). The activated particle number concentration levels off with higher SS % as particles reach 100% activation. These plots were generated using the ARM Facility Data Quality Diagnostic Plot Browser (http://plot.dmf.arm.gov/plotbrowser/).





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Figure 2. Aerosol size distribution as measured on September 8, 2015 by the CCN-200 deployed during the Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) field campaign, Azores, Portugal.

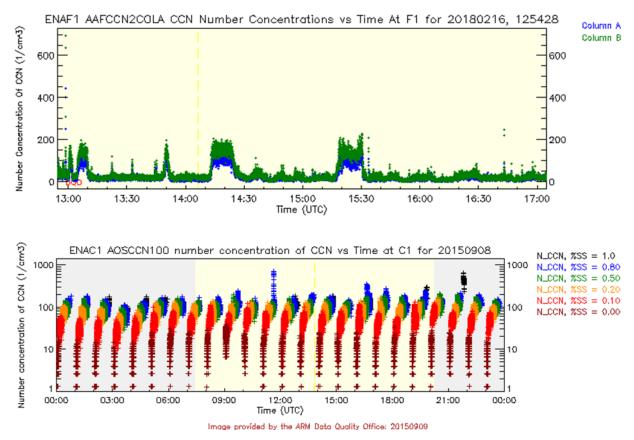


Figure 3. Total aerosol particle number concentration as measured by the CCN-200 on February 16, 2016 during the ACE-ENA field campaign, Azores, Portugal.

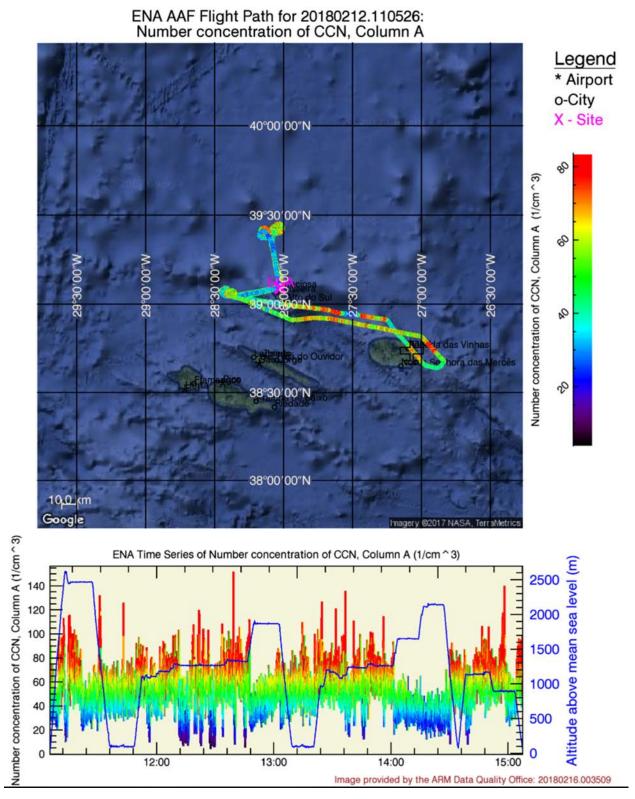


Figure 4. The flight path plot colored by the total aerosol particle number concentration measured by the column A of the CCN-200 on February 12, 2016 during the ACE-ENA field campaign, Azores, Portugal.

6.4 Data Quality

Data quality evaluation involves automatic flagging of data based on criteria developed by instrument mentors and automatic generation of plots in collaboration with the ARM Data Quality Office.

Automatic data quality checks include:

- Checking that sheath/sample flow rate ratio is between 9.5 and 10.5.
- Checking that the OPC first-stage voltage monitor reading is below 0.5 volts (V) (verifying proper operation of the OPC).

Plots that are automatically generated by the ARM Data Quality Office include (see also Figure 2 and Figure 3):

- Aerosol particle size distribution as a function of time. Low counts or noisy signal may indicate issues with the OPC.
- Total number concentration of humidified particles as a function of time. Lack of clear step-wise change in particle concentration with changing humidifier supersaturation may indicate an issue with the humidifier column.
- Sample flow rate. Low or unstable flow rate indicates a blockage in the sample line or a failing pump.
- Laser current and reference voltage. High current and reference voltage indicate a failing OPC.

6.5 Calibration Database

During deployment the CCN is periodically calibrated by instrument mentors. CCN calibration involves generating and size-selecting ammonium sulfate particles and recording their total number concentration before and after activation in the CCN as a function of particle size. This is done for several humidifier column temperature gradients. Next, the 50% activation diameter (particle diameter where 50% of the generated ammonium sulfate particles are activated) is calculated for each humidifier column temperature gradient. Because the activation characteristics of ammonium sulfate are known (2), supersaturation % can be calculated from the 50% activation diameter to establish/verify the relation between the temperature gradient of the humidifier column and the supersaturation %.

Mentor-provided calibrations are used to calculate the thermal efficiency (eta) of the CCN column. Based on the eta and measured (not set-point) thermal gradients, supersaturation values are updated during the data ingest. This real-time calibration approach allows mentors to correct for pressure, flows, and supersaturation drifts. Furthermore, monitoring long-term eta values allows assessment of the overall performance of the CCN column. Coefficients are also recorded in the ARM Operation Status System (OSS, <u>https://oss.arm.gov/oss.php</u>) and archived by the instrument mentors.

7.0 Technical Specification

7.1 Units

Aerosol particle size: micrometers (μ m); aerosol particle number concentration: particles per cubic centimeter (cm-3) or raw number of counts (dimensionless); supersaturation: % (dimensionless).

7.2 Range

Supersaturation can be varied from 0.07% to 2.0%. Particle size (after humidification) can be measured between 0.75-10 μ m.

The particle number concentration measurement range depends on the supersaturation (due to growth kinetics of activated particles). At SS below 0.2% the maximum particle number concentration is 6000 s- 1 and at SS above 0.3% it is 20000 s-1.

7.3 Accuracy

Accuracy of supersaturation control depends on the accuracy of pressure, flow, and temperature sensors. At supersaturations above 0.1% the accuracy is within 3% (2). Below 0.1%, the supersaturation dependence on the humidifier column temperature gradient becomes non-linear, and unless taken into account in instrument calibration and data interpretation (by default it is not), accuracy of supersaturation can be as low as 40%.

Accuracy of single particle counting depends on the total aerosol particle concentration and is within 4% for the operating ranges specified above.

7.4 Repeatability

Repeatability of supersaturation control depends on the stability of flow and temperature control. In laboratory conditions the repeatability of supersaturation control is $\pm 1\%$. In the field, this increases to $\pm 5\%$ due to fluctuations of ambient temperature (2).

7.5 Sensitivity

Aerosol particle size and concentration measurement are sensitive to particle concentration (due to particle coincidence during counting at higher concentrations). Growth kinetics of the particles in the humidifier column set the upper limit of total particle counts for accurate single-particle detection at 6000 s-1 for supersaturations below 0.2% and at 20000 s-1 for supersaturations above 0.3%.

The relationship between humidifier column temperature gradient and supersaturation % is sensitive to ambient pressure. For accurate measurements, coefficients from a calibration done at similar conditions (altitude) as the measurement location should be used.

7.6 Uncertainty

Uncertainty of activated particle sizing is largely determined by the sizing resolution of the OPC and is approximately $\pm 0.25 \ \mu m$.

7.7 Input Values

Parameters set by the user include:

Supersaturation scanning schedule: desired supersaturations and their durations, sample- and sheath flow rates.

The supersaturation scanning schedule is typically chosen to fit a particular measurement location. This is because the properties of aerosol particles can vary by location and reach 100% activation at different supersaturation % values.

7.8 Output Values

The recorded data include:

Measurement date/time, supersaturation, humidifier temperatures, system temperatures, sample flow, sheath flow, sample pressure, laser current, OPC monitor voltages, particle number concentration by size bin (20 bins), and total particle number concentration.

8.0 Instrument System Functional Diagram

The entire CCN system can be divided into several subsystems:

- 1. Flow system. Brings the sample aerosol through the humidifier column to the optical particle counter (OPC); controls and measures the flows.
- 2. Humidifier. Controls the water flow and internal temperatures of the humidifier column.
- 3. Control electronics. Includes the data acquisition and control boards and a built-in personal computer (PC) running a version of the Microsoft Windows operating system.

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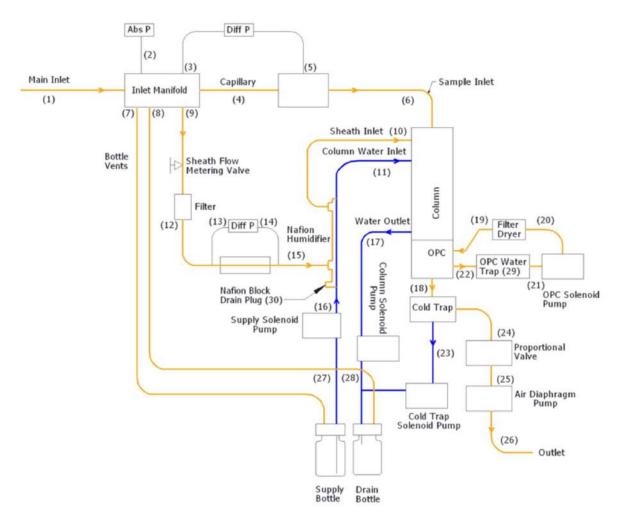


Figure 5. Schematic diagram of the CCN (CCN-100) flow system. Adapted from the manufacturer's manual.

9.0 Instrument/Measurement Theory

The CCN counter is a continuous-flow, thermal-gradient diffusion chamber for measuring aerosols that can act as cloud condensation nuclei (1). The CCN draws an aerosol sample into a column, where a thermodynamically unstable, supersaturated water vapor condition is created by taking advantage of the difference in diffusion rates between water vapor and heat. Water vapor diffuses from the warm, wet column walls toward the centerline at a faster rate than the heat. The wall temperature along the column gradually increases to create a well-controlled and quasi-uniform centerline supersaturation. Seeking equilibrium, the supersaturated water vapor condenses on the cloud condensation nuclei in the sample air to form droplets, just as cloud drops form in the atmosphere. An OPC using side-scattering technology counts and sizes the activated droplets.

10.0 Set-Up and Operation of Instrument

- 1. Connect a keyboard and a mouse to the universal serial bus (USB) ports and an external computer display to the video graphics array (VGA) port on the instrument (for initial set-up only).
- 2. Connect the external CCN power supply (for initial set-up only).
- 3. Install the water supply and drain bottles (for initial set-up only). Make sure the supply bottle is filled with distilled water (tap water or any purified water with added minerals is not OK) and the drain bottle is empty.
- 4. Switch on the instrument and wait for the internal computer to boot up. The measurement software is configured to start automatically.
- 5. If the CCN is being started for the first time after transport and its system does not contain water, a dry startup procedure must be performed. During the first 20 seconds after the program starts, click the Dry Start Up button shown in the "SS settings" tab of the software main window (the Dry Start Up and Dry Shut Down buttons disappear after 20 seconds). Selecting Dry Start Up will set the liquid supply pump to high and disable the CCN concentration alarm.
- 6. When the CCN humidifier column is fully wetted, the OPC should be counting (note that with dry startup it may take 4 to 12 hours for the CCN to become properly humidified and count particles). At this time, the status light in the upper center of the window should be green. A green status light indicates the dual CCN instrument is functioning properly.
- 7. You can now begin to collect data.

11.0 Software

Instrument control and data acquisition is performed by NI LabView-based software written by the manufacturer. Additional LabView-based software, written by Brookhaven National Laboratory, reformats and relocates the data files saved by the manufacturer's software.

12.0 Calibration

The CCN is calibrated by the manufacturer before delivery to the user and during instrument maintenance at the manufacturer's facilities. The instrument mentors also typically perform calibration before and after each deployment at conditions (altitude) similar to the measurement site and during deployment if it has been more than a year from the last calibration and the deployment is not yet ending. The calibration schedule is flexible because it depends on the availability of the calibration equipment (calibration scanning mobility particle sizer spectrometer [SMPS]).

Mentor-provided calibrations are used to calculate the thermal efficiency (eta) of the CCN column. Based on the eta and measured (not set-point) thermal gradients, supersaturation values are updated during the data ingest. This real-time calibration approach allows mentors to correct for pressure, flows, and supersaturation drifts. Calibration results are also used by the mentors to assess the overall condition of the instrument, especially the humidifier column.

Manufacturer's calibration includes:

- Supersaturation calibration with ammonium sulfate aerosol particles (see below).
- Size calibration of the OPC with National Institute of Standards and Technology (NIST)-traceable polystyrene latex (PSL) particles.
- Flow calibrations with a precision flow meter.

Mentor calibration of the CCN involves generating and size-selecting ammonium sulfate particles and recording their total number concentration before and after activation in the CCN as a function of particle size. This is done for several humidifier column temperature gradients. Next, the 50% activation diameter (particle diameter where 50% of the generated ammonium sulfate particles are activated) is calculated for each humidifier column temperature gradient. Because the activation characteristics of ammonium sulfate are known (2), supersaturation % can be calculated from the 50% activation diameter to establish/verify the relationship between the temperature gradient of the humidifier column and the supersaturation %.

13.0 Maintenance

Action when:

- Adding distilled water to the CCN fill bottle (every day).
- Emptying the CCN drain bottle (every time the fill bottle is filled). Inspect the waste water for presence of green algae. Clean the CCN water system if algae is present.
- Emptying the CCN water trap bottles (as needed, when water is present).

14.0 Safety

The CCN-100 and CCN-200 are Class IIIb Laser Products. During normal operation, the user is not exposed to laser radiation.

15.0 Citable References

(1) Roberts, GC, and Nenes, A. 2005. "A Continuous-Flow Streamwise Thermal-Gradient CCN Chamber for Atmospheric Measurements." *Aerosol Science and Technology 39*(3): 206–221, http://doi.org/10.1080/027868290913988

(2) Rose, D, GP Frank, U Dusek, SS Gunthe, MO Andreae, and U Pöschl, 2007. "Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment." *Atmospheric Chemistry and Physics Discussions* 7(3): 8193–8260, <u>http://doi.org/10.5194/acpd-7-8193-2007</u>

(3) Raatikainen, T, JJ Lin, KM Cerully, TL Lathem, RH Moore, and A Nenes. 2014. "CCN data interpretation under dynamic operation conditions." *Aerosol Science and Technology* 48(5): 552–561, https://doi.org/10.1080/02786826.2014.899429



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