

Cloud Condensation Nuclei Hygroscopicity Value-Added Product Report

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Acronyms and Abbreviations

ARM	Atmospheric Radiation Measurement
CCN	cloud condensation nuclei
CCNC	cloud condensation nuclei counter
CPC	condensation particle counter
NetCDF	Network Common Data Form
SMPS	scanning mobility particle sizer
SGP	Southern Great Plains
SS	supersaturation
UTC	Coordinated Universal Time
VAP	value-added product

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1.0 Introduction

The purpose of the Atmospheric Radiation Measurement (ARM) user facility's cloud condensation nuclei hygroscopicity parameter (AOSCCNSMPSKAPPA) value-added product (VAP) is to calculate the hygroscopicity parameter, κ , to quantify the ability of aerosols to activate into cloud water droplets. The hygroscopicity parameter is often used to model the cloud condensation nuclei (CCN) activity of atmospheric aerosols of different sizes and compositions, providing additional insight on the influence of aerosols on climate.

Laboratory experiments show that the κ values for highly hygroscopic aerosols, such as salts and sulfates, vary from 0.5 to 1.4 (Petters and Kreidenweis 2007). For organic compounds they are observed to vary between 0.01 and 0.5. For non-hygroscopic aerosols, such as soot, κ values are very close to zero. Ambient aerosols are complex mixtures of organic and inorganic compounds and previous observations indicate that κ values typically vary from 0.05 to 0.9 (Petters and Kreidenweis 2007).

2.0 Algorithm and Methodology

To calculate κ (κ), this VAP draws on collocated aerosol particle size data from the scanning mobility particle sizer (SMPS) and aggregated CCN number concentrations from the cloud condensation nuclei counter (CCNC) datastreams (CCN2COLAAVG) managed by the ARM facility.

κ -Köhler theory (Petters and Kreidenweis 2007) is used to calculate κ from aerosol particle size and supersaturation measurements. In these calculations, the influence of mixing state and individual chemical composition of particles are ignored, and the overall κ is calculated based on the particle number size distribution from SMPS and CCN concentrations at a defined instrument supersaturation as a function of time, assuming particles are internally mixed with bulk chemical composition. The backward stepwise integration from the upper size limit of the SMPS measurements is performed until the total particle concentration matches with the measured CCN concentrations. The corresponding particle diameter where SMPS and CCNC concentrations are equal is assumed to be the critical diameter required for activation. The particles greater than this critical diameter are assumed to be those particles that activate into the CCN (e.g., Ren et al. 2018). At each value of supersaturation measured by the instrument, this critical diameter is then used to calculate the κ .

A single parameter representation of the hygroscopicity of aerosol particles (or κ) is described by Petters and Kreidenweis (2007). The critical water vapor supersaturation (S_c) that a particle must be exposed to in order to activate as a CCN is given by Equation (1), which can be also rearranged to calculate the κ (Equation 2).

$$S_c = \left[\frac{4}{\kappa D_p^3} \left(\frac{4\sigma M_w}{3RT\rho_w} \right)^3 \right]^{1/2} \quad (1)$$

$$\kappa = \frac{4}{27D_p^3 \ln^2 S_c} \left(\frac{4\sigma M_w}{3RT \rho_w} \right)^3 \quad (2)$$

Here, κ refers to the particle hygroscopicity (kappa) parameter, D_p is the dry particle diameter, M_w is the molar mass of water, ρ_w is the density of water, and σ is the droplet surface tension at activation.

In our calculations, we assume the surface tension for all droplets is 0.072 Jm^{-2} . It is important to note that our methodology also assumes that all particles have uniform chemical composition and mixing state. As previously mentioned, we also assume that particles with larger diameters preferentially activate into water droplets over particles with smaller diameters independent of their chemical composition at a given value of supersaturation (Dusek et al. 2006). Due to these assumptions, the κ value derived from this VAP will depend on instrument supersaturation and critical diameter. In principle, aerosol particles of constant chemical composition have identical κ , regardless of the particle diameter and the instrument saturation at which measurements are taken. Using this analysis method, a polydisperse size distribution of a single-component aerosol particle measured with the CCN instrument would appear to have higher κ at lower instrument supersaturation. This method works best when used to calculate the κ of aerosol from clean continental regions/sites due to lesser heterogeneity in their chemical composition across the size distribution.

3.0 Input Data

The AOSCCNSMPSKAPPA VAP has two input datastreams: `aosccn2colaavg.b1` and `aoスマps.b1`. These datastreams are drawn from the SMPS and CCNC ARM instruments as shown in Figure 1. The SMPS measures the number size distribution of ambient aerosols in the range of 10-515 nm. The SMPS is operated in a scanning mode from a 10-nm diameter stepped upwards to 515 nm for a total of 109 diameter steps, such that a complete scan was completed in 300 seconds. The CCNC is operated at various water vapor supersaturation (SS) values by regulating the temperature gradient between the top and bottom wetted column of the counter. The SS values are typically varied from 0.0 to 1.0 % stepping upwards at each single SS step: 0, 0.1, 0.2, 0.4, 0.8, and 1% for 10 minutes. A complete CCN scan takes 60 minutes. Figure 2 depicts the data flow from the SMPS and CCN instruments into the input datastreams used by this VAP. Information about the quality checks applied in the `aosccn2colaavg.b1` datastream can be found in the Appendix of this report. For more information about the input data to this product, see the SMPS and CCN instrument handbooks (Kuang 2016, Uin 2016).

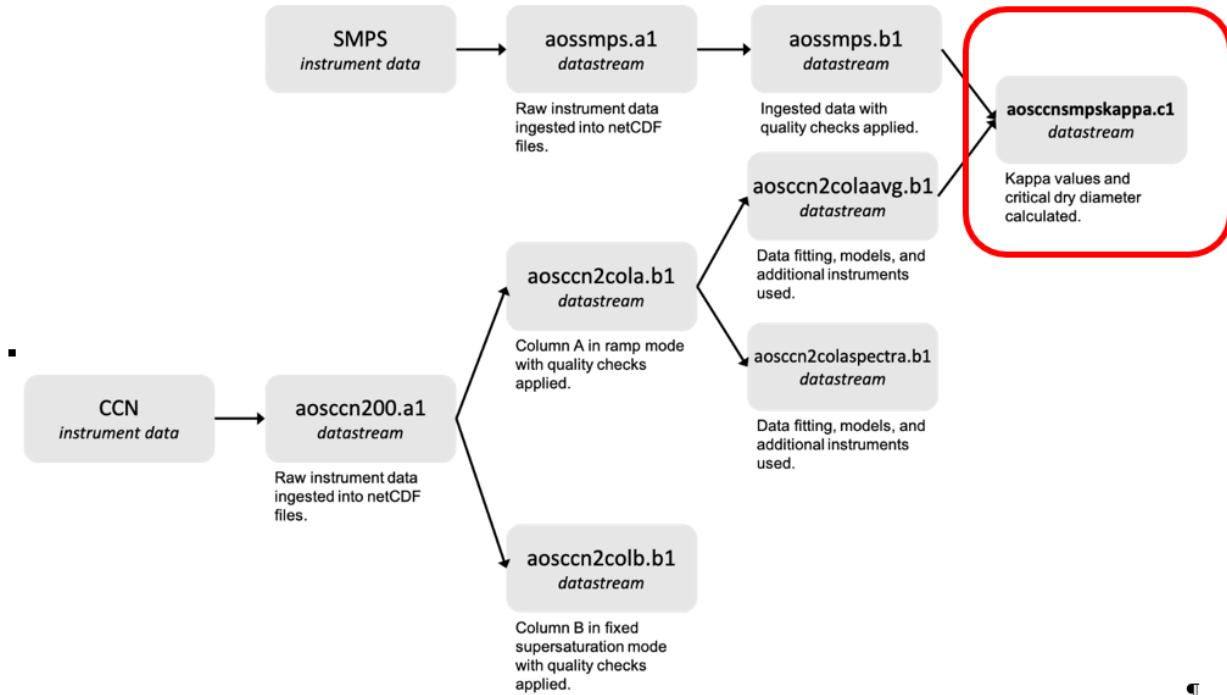


Figure 1. Diagram depicting data flows for the SMPS and CCN instruments. The highlighted box shows the VAP product datastream described in this report.

The aosccnsmplskappa.c1 VAP retrieves a selection of variables from the aossmpls.b1 and aosccn2colaavg.b1 datastreams. The variables retrieved from each input datastream are listed below in Table 1 and Table 2.

Table 1. Variables retrieved from the aosccn2colaavg.b1 input datastream.

Variable Name	Description
aerosol_number_concentration	Aerosol particle number concentration derived from condensation particle counter (CPC) data. Ambient aerosol particles are dried before size-distribution measurements are performed.
alt	Altitude above mean sea level.
droplet_size	Size bins for cloud condensation nuclei droplets.
droplet_size_bounds	Droplet size bin bounds.
lat	North latitude.
lon	East longitude.
N_CCN	Mean number concentration of ambient aerosol particles in air.
N_CCN_dN	Droplet count by bin size.

Variable Name	Description
qc_N_CCN	Quality check results on field: Mean number concentration of N_CCN.
setpoint	Supersaturation set point bins.
supersaturation_calculated	Mean calculated supersaturation values for a fixed supersaturation set point.

Table 2. Variables retrieved from the aoassmps.b1 input datastream.

Variable Name	Description
diameter_mobility	Midpoint of geometric mean mobility diameter.
diameter_mobility_bounds	Mobility diameter bin boundaries.
dN_dlogDp	The aerosol number size distribution where the number of particles per bin (dN) has been divided by the bin-width in log10 space (dlogDp). This simplifies comparison of size distributions from instruments with different bin spacing.
qc_dN_dlogDp	Quality check results on field: Number size distribution, electrical mobility diameter.

4.0 Output Data

The AOSCCNSMPSKAPPA VAP produces one output datastream: aosccnsmpskappa.c1. Output files are produced daily and named as XXXaosccnsmpskappaYY.c1.YYYYMMDD.HHMMSS.nc, where XXX is the site where the VAP is running, YY is the facility designation within the site, YYYYMMDD is the date of the first point in the file, and HHMMSS is the time of the first point in the file.

Each output file contains all the input variables described by the tables in §3 in addition to the following calculated variables.

Table 3. Variables calculated by the aosccnsmpskappa.c1 VAP.

Variable Name	Description
critical_diameter	The critical dry diameter used to calculate κ .
kappa	CCN derived hygroscopicity parameter kappa (κ), calculated using κ -Köhler theory with fixed thermodynamic constants.

This VAP also produces several plots designed to provide a quick overview of the data produced on a daily basis. A selection of these plots is listed below.

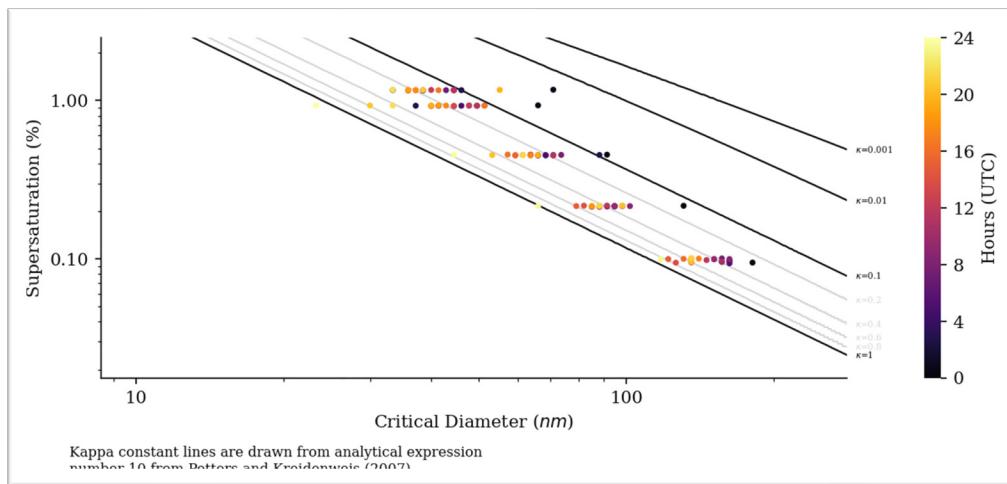


Figure 2. Supersaturation versus critical diameter with reference κ constant lines on 2020-06-27.

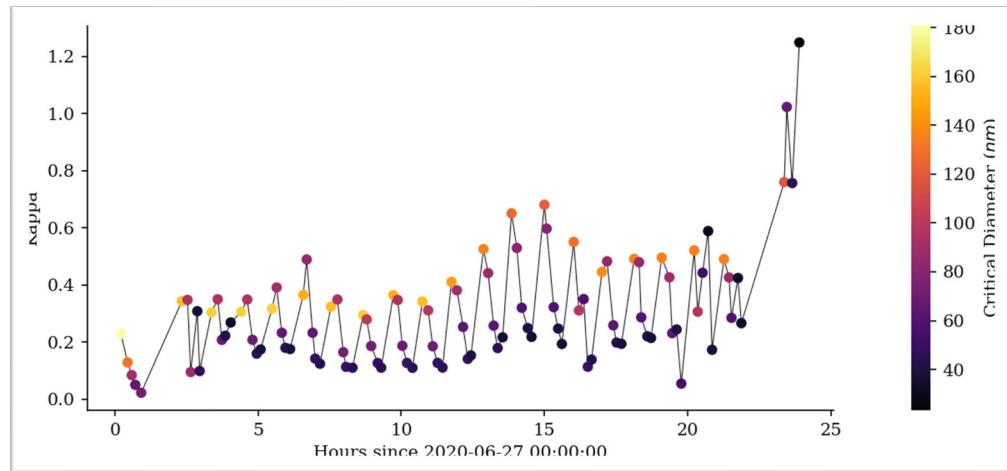


Figure 3. κ versus time on 2020-06-27.

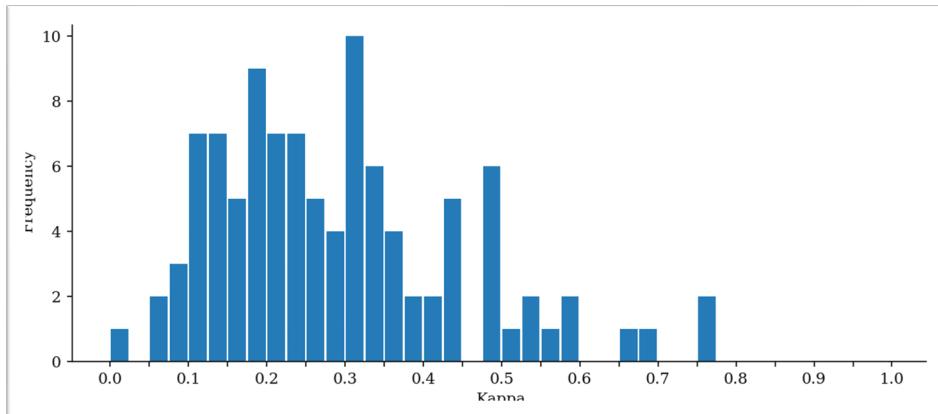


Figure 4. Frequency distribution of κ on 2020-06-27.

5.0 Summary

The AOSCCNSMPSKAPPA VAP provides hygroscopicity parameter kappa calculations that can be used to characterize aerosol properties at ARM sites and facilities. The VAP produces daily netCDF files, each of which spans a 24-hour interval beginning at midnight (UTC). The temporal spacing between data points in each file can vary due to the different time-grids used by the CCN and SMPS input datastreams but is typically around five minutes. A several-day delay on processing is imposed to allow input data to become fully available before the VAP is run.

This κ product allows us to predict the CCN properties of ambient aerosol from clean continental or remote sites. The reported $\kappa - S_c$ relationship can be used to derive the CCN spectrum from aerosol size-distribution to perform aerosol-CCN closure studies to further understand the CCN impacts on warm clouds.

6.0 References

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Appendix A

Quality Checks Applied in aosccn2cola.b1 Datastream

Figure 5 shows a plot of the aosccn2cola.b1 cloud condensation nuclei particle counts (N_CCN), including quality assessments assigned to the data at each timestamp shown. The (N_CCN) concentrations marked as good are shown in green and are used to develop the AVG and SPECTRA higher-level data products shown in Figure 1. Figure 5 was generated using the ARM_nc_display Matlab function (courtesy of Connor Flynn).

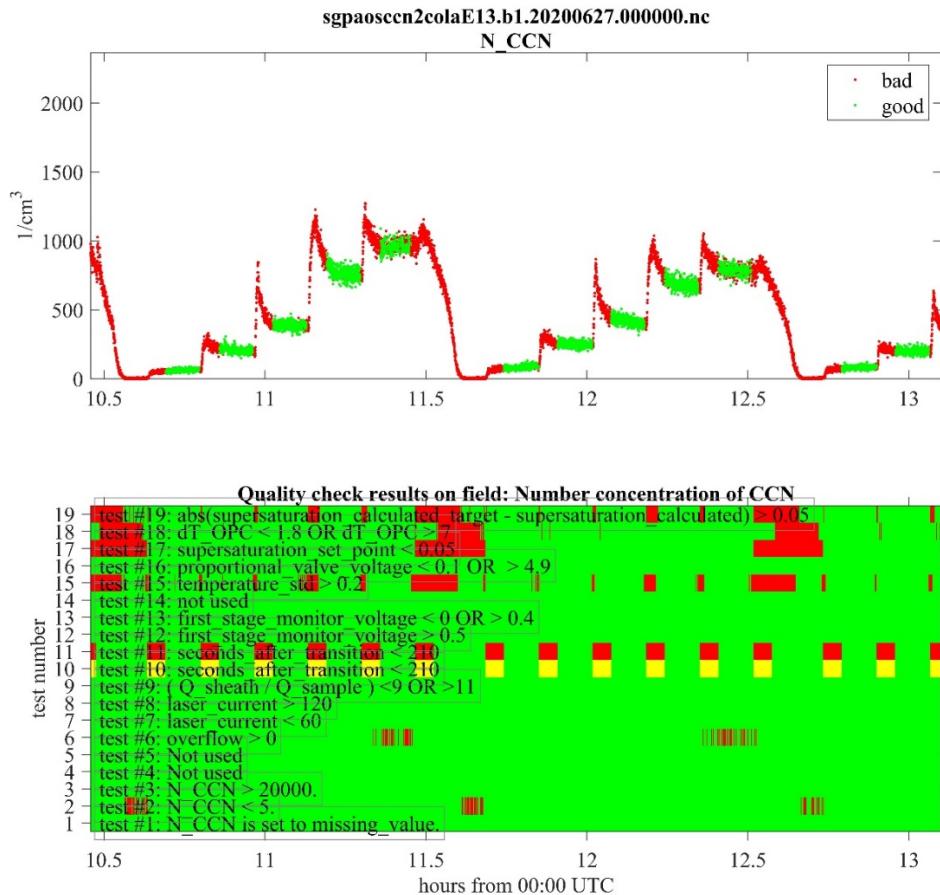


Figure 5. CCN concentrations and quality assessments from aosccn2cola.b1 at the Southern Great Plains (SGP) Extended Facility 13 (E13).



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