U.S. DEPARTMENT OF ENERGY | Office of Science

DOE/SC-ARM-TR-272

Cloud Condensation Nuclei Hygroscopicity Value-Added Product Report

G Kulkarni
JE Shilling
MS Levin

November 2023

DOI/SC-ARM-TR-272

Cloud Condensation Nuclei Hygroscopicity Value-Added Product Report

G Kulkarni
MS Levin
JE Shilling
All at Pacific Northwest National Laboratory

November 2023

How to cite this document:

Kulkarni, G, MS Levin, and J Shilling. 2023. Cloud Condensation Nuclei Hygroscopicity Value Added Product Report. U.S. Department of Energy, Atmospheric Radiation Measurement user facility, Richland, Washington. DOE/SC-ARM-TR-272.

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

Acronyms and Abbreviations

ARM	Atmospheric Radiation Measurement
CCN	cloud condensation nuclei
CCNC	cloud condensation nuclei counter
CPC	condensation particle counter
ENA	Eastern North Atlantic
netCDF	Network Common Data Form
SGP	Southern Great Plains
SMPS	scanning mobility particle sizer
SS	supersaturation
UHSAS	ultra-high-sensitivity aerosol spectrometer
UTC	Coordinated Universal Time
VAP	value-added product

Contents

Acronyms and Abbreviations iii
1.0 Introduction 1
2.0 Algorithm and Methodology 1
    2.1 AOSCCNSMPSKAPPA 1
    2.2 AOSCCNUHSASKAPPA 2
3.0 Input Data 2
    3.1 AOSCCNSMPSKAPPA 2
    3.2 ASCCNUHSASKAPPA 5
4.0 Output Data 5
    4.1 AOSCCNSMPSKAPPA 5
    4.2 AOSCCNUHSASKAPPA 7
5.0 Summary 10
6.0 References 10
Appendix A – Quality Checks Applied in aosccn2cola.b1 Datastream A.1

Figures

1 Diagram depicting data flows for the SMPS and CCN instruments 3
2 Supersaturation versus critical diameter with reference κ constant lines on 2020-06-27 at the Southern Great Plains (SGP) E13 observatory using SMPS data. 4
3 κ versus time on 2020-06-27 at the SGP E13 observatory using SMPS data. 6
4 Frequency distribution of κ on 2020-06-27 at the SGP E13 observatory using SMPS data. 7
5 Supersaturation versus critical diameter with reference κ constant lines on 2016-08-01 at the Eastern North Atlantic (ENA) C1 observatory using UHSAS data. 8
6 κ versus time on 2016-08-01 at the ENA C1 observatory using UHSAS data. 9
7 Frequency distribution of κ on 2016-08-01 using UHSAS data. 9
8 CCN concentrations and quality assessments from aosccn2cola.b1 at SGP E13. .... A.1

Tables

1 Variables retrieved from the aosccn2colaavg.b1 input datastream. 4
2 Variables retrieved from the aossmps.b1 input datastream. 5
3 Variables retrieved from the aosuhsas.b1 input datastream. 5
4 Variables calculated by the aosccnsmpskappa.c1 VAP. 6

1.0 Introduction

The purpose of the U.S. Department of Energy Atmospheric Radiation Measurement (ARM) user facility's Cloud Condensation Nuclei Hygroscopicity Parameter (AOSCCNSMPSKAPPA and AOSCCNUHSASKAPPA) Value-Added Product (VAP) is to calculate the hygroscopicity parameter, kappa, 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. The AOSCCNUHSASKAPPA VAP was recently developed to provides kappa data for ARM sites where AOSCCNSMPSKAPPA was missing.

Laboratory experiments show that the kappa values for highly hygroscopic aerosols 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, kappa values are very close to zero. Ambient aerosols are complex mixtures of organic and inorganic compounds and previous observations indicate that kappa values for these aerosols typically vary from 0.05 to 0.9 (Petters and Kreidenweis 2007).

2.0 Algorithm and Methodology

2.1 AOSCCNSMPSKAPPA

To calculate kappa (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 (CCN2COLAAVG) datastreams 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 is 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 the measured CCN concentrations. The corresponding particle diameter where integrated 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 to activate as a CCN is given by Equation 1, which can be also rearranged to calculate the κ (Equation 2).

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

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

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 that of water (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 single-component aerosol particles measured with the CCN instrument would appear to have higher κ at lower instrument supersaturation, an obvious error. 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.

2.2 AOSCCNUHSASKAPPA

The AOSCCNUHSASKAPPA kappa (κ) VAP product was developed using similar methodology to that described in section 2.1 but using collocated aerosol particle size data from the ultra-high-sensitivity aerosol spectrometer (UHSAS) instead of the SMPS instrument. The UHSAS detects particle sizes from 60 to 1000 nm, and the aerosol size distribution data is recorded every 10 seconds (Uin 2016). Due to uncertainty at the lower and upper detection limits of the instrument, only the particles in the size range 70 to 700 nm were used in the kappa calculation. The lower limit constraint on the particle size restricted the calculation of kappa to supersaturation values below 0.5% (S_c< 0.5 %) because, for larger S_c values, the critical diameter was often less than 70 nm.

3.0 Input Data

3.1 AOSCCNSMPSKAPPA

The AOSCCNSMPSKAPPA VAP has two input datastreams: aosccn2colaavg.b1 and aossmps.b1. These datastreams are drawn from the SMPS and cloud condensation nuclei counter (CCNC) 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 across this size range and data is distributed into 109 size bins with a complete scan 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% singling six SS steps: 0, 0.1, 0.2, 0.4, 0.8, and 1% with data recorded at each SS value for10 minutes. A complete CCN scan cycle 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, please see the SMPS and CCN technical reports (Kuang 2016, Uin 2022).

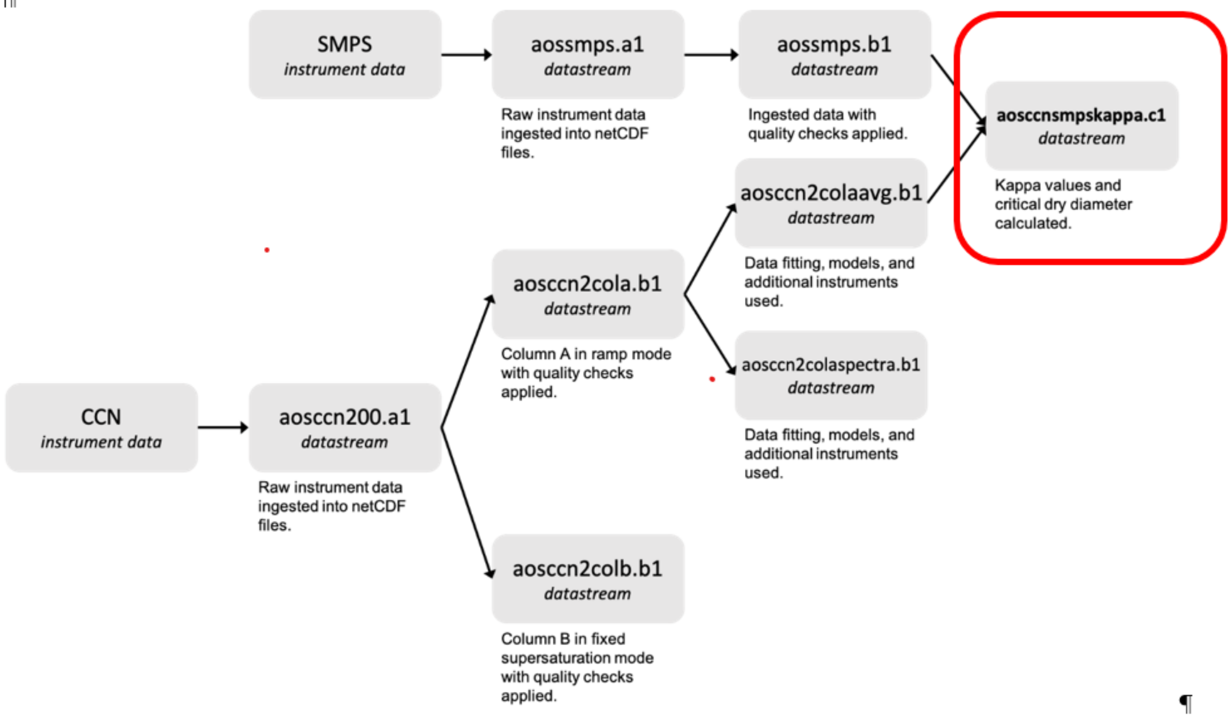

Figure 1. Diagram depicting data flows for the SMPS and CCN instruments. The highlighted box shows the VAP product datastream described in this report.

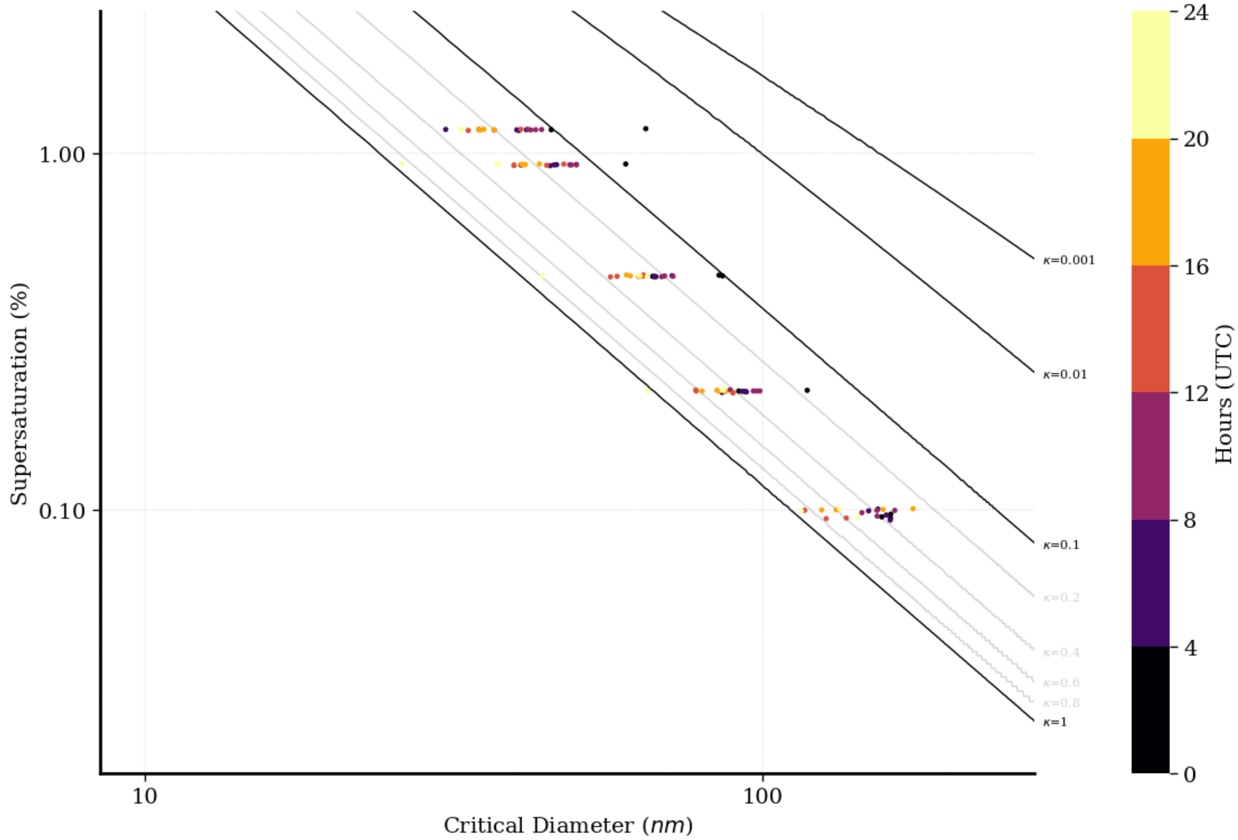

Figure 2. Supersaturation versus critical diameter with reference κ constant lines on 2020-06-27 at the Southern Great Plains (SGP) E13 observatory using SMPS data.

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

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

Variable Name	Description
aerosol_number_concentration	Aerosol particle number concentration derived from CPC data. Ambient aerosol particles are dried before size-distribution measurements are performed.
droplet_size	Size bins for cloud condensation nuclei droplets.
droplet_size_bounds	Droplet size bin bounds.
N_CCN	Mean number concentration of ambient aerosol particles in air.
N_CCN_dN	Droplet count by bin size.
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 aossmps.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.

3.2 ASCCNUHSASKAPPA

The AOSCCNUHSASKAPPA kappa (κ) VAP product uses UHSAS instrument data instead of SMPS data (Figure 1). The aosuhsas.b1 and aosccn2colaavg.b1 datastreams are used. The variables retrieved from aosuhsas.b1 datastream used in this VAP are listed below in Table 3.

Table 3. Variables retrieved from the aosuhsas.b1 input datastream.

Variable Name	Description
diameter_optical	Midpoint of geometric mean optical diameter
Diameter_optical_bounds	Optical diameter bin boundaries.
dN_dlogDp	Number size distribution, optical scattering diameter at 1054 nm. 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, optical diameter (70 to 700 nm).
qc_total_N_conc	Quality check results on field: Total number concentration from integrated size distribution (< 3600 #/cc)

4.0 Output Data

4.1 AOSCCNSMPSKAPPA

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 Section 3 in addition to the calculated variables in Table 4.

Table 4. 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 daily. A selection of these plots is provided in Figures 3 and 4.

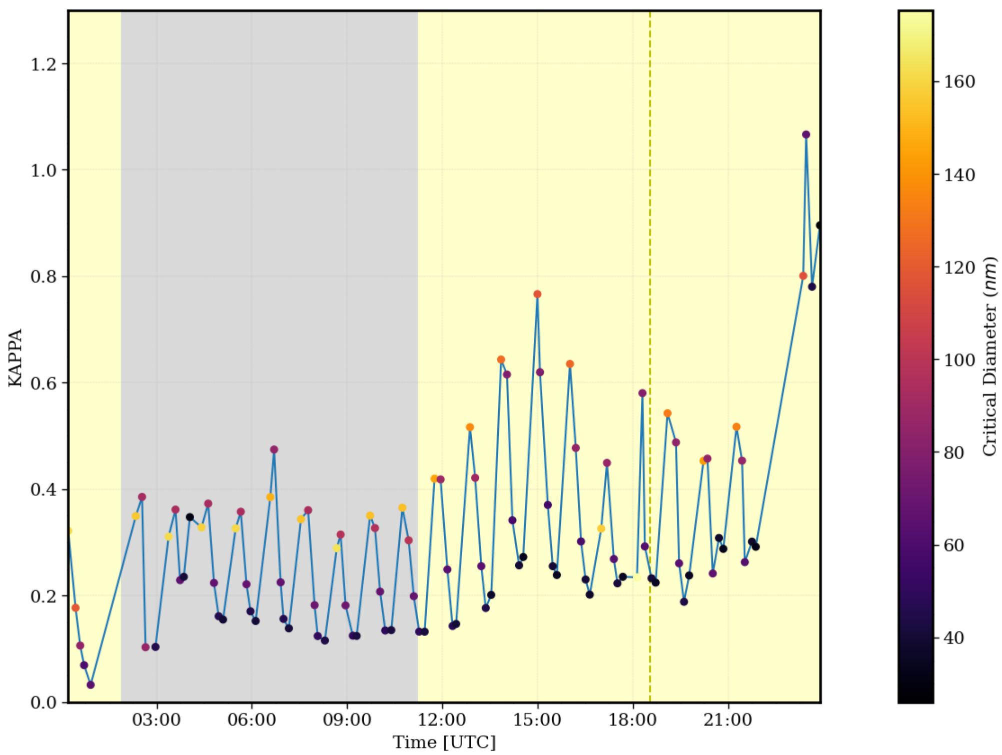

Figure 3. κ versus time on 2020-06-27 at the SGP E13 observatory using SMPS data.

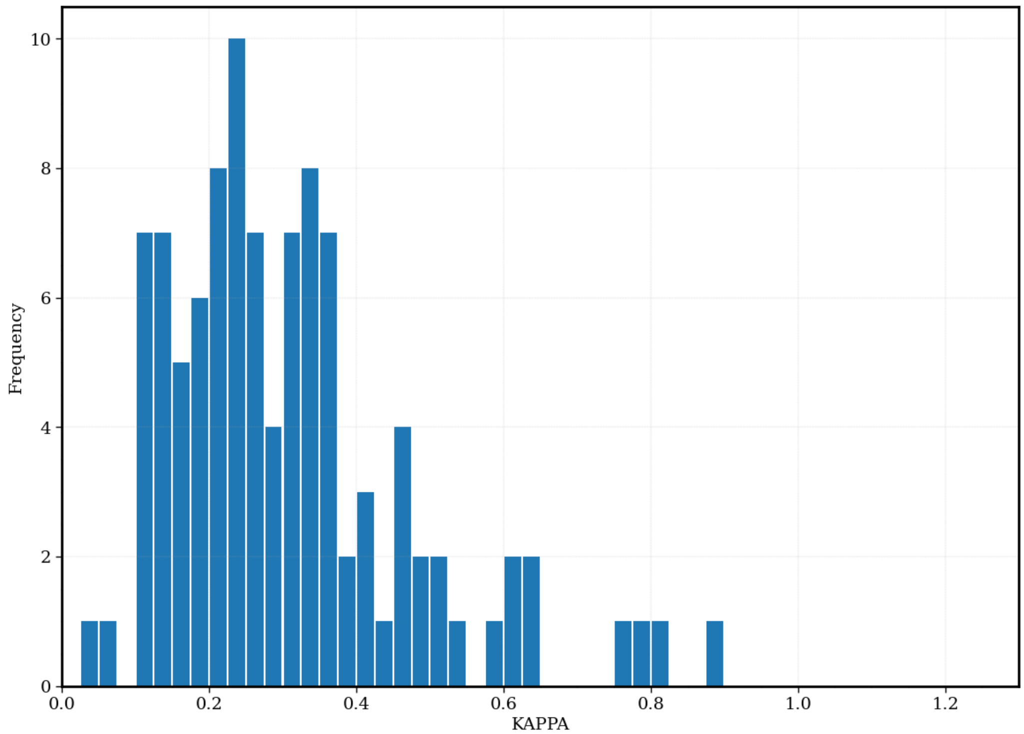

Figure 4. Frequency distribution of κ on 2020-06-27 at the SGP E13 observatory using SMPS data.

4.2 AOSCCNUHSASKAPPA

The AOSCCNUHSASKAPPA VAP produces one output datastream: aosccnuhsaskappa.c1. Output files are produced daily and named as XXXaosccnuhsaskappaYY.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.

The following output can be obtained, as shown in Figures 5–7.

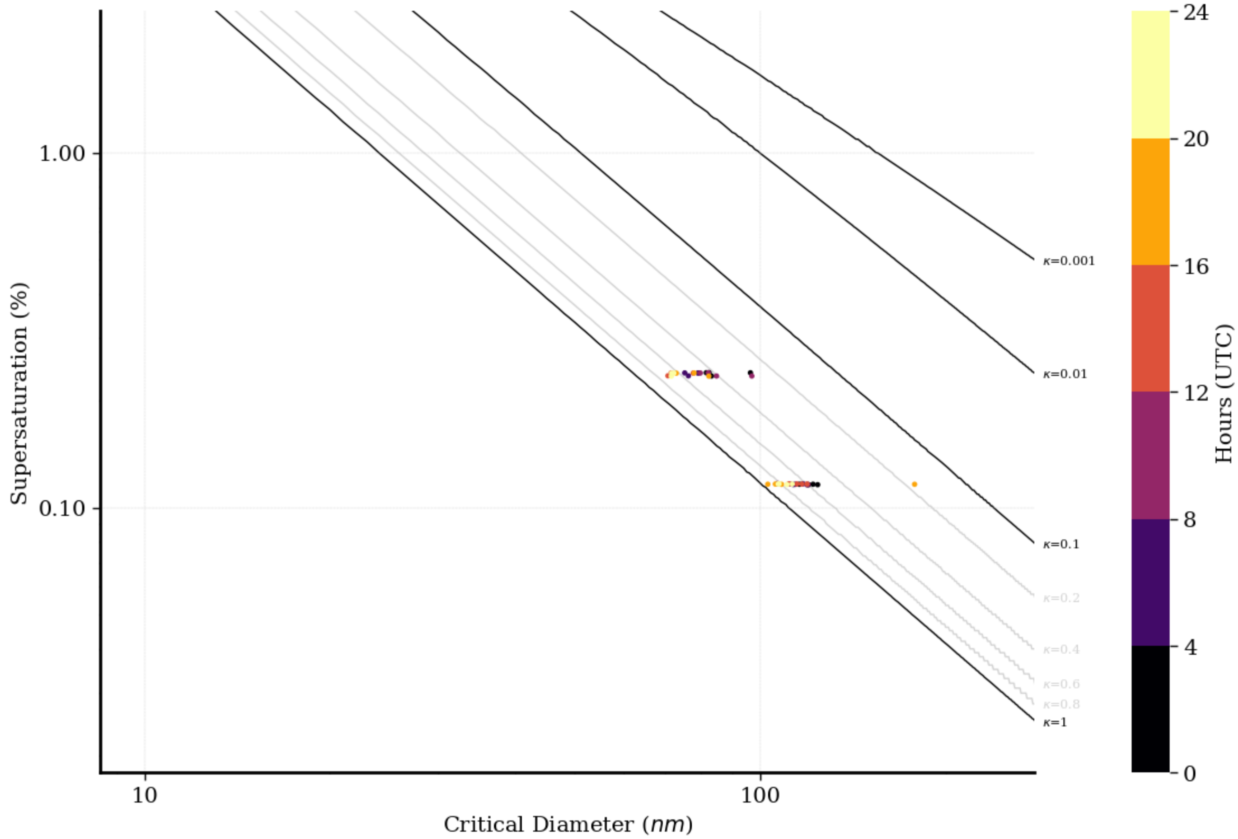

Figure 5. Supersaturation versus critical diameter with reference κ constant lines on 2016-08-01 at the Eastern North Atlantic (ENA) C1 observatory using UHSAS data.

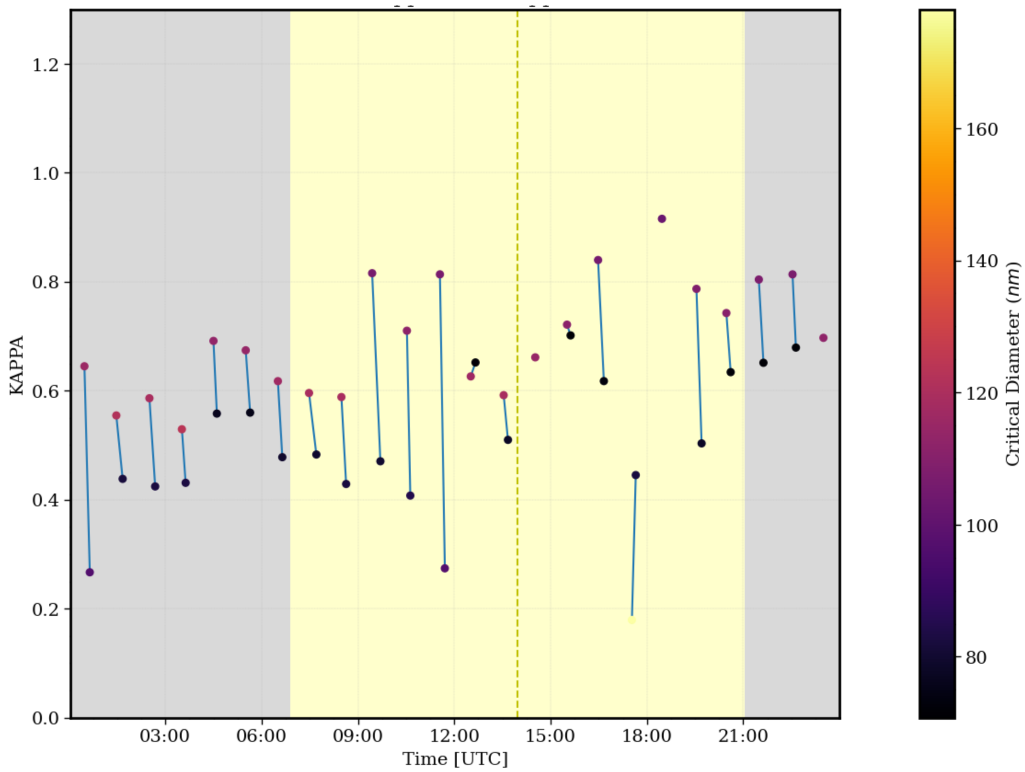

Figure 6. κ versus time on 2016-08-01 at the ENA C1 observatory using UHSAS data.

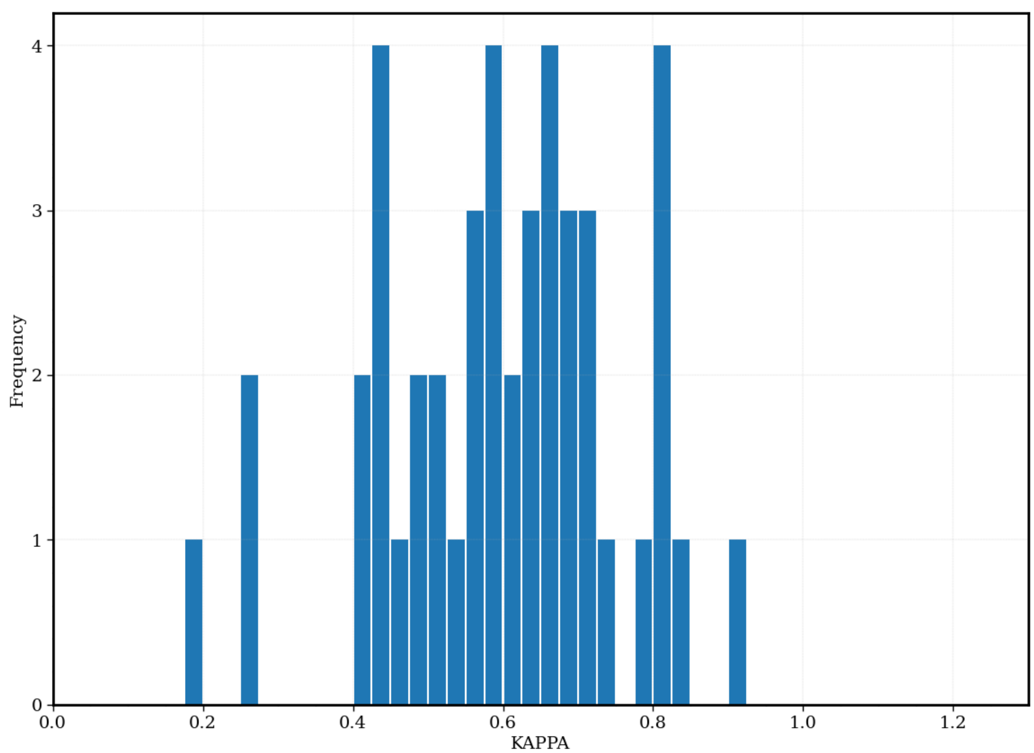

Figure 7. Frequency distribution of κ on 2016-08-01 using UHSAS data.

5.0 Summary

The two AOSCCNSMPSKAPPA and AOSCCNUHSASKAPPA VAP products provide hygroscopicity parameter (kappa) calculations that can be used to characterize aerosol properties at ARM observatories and mobile 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/UHSAS input datastreams. A several-day delay on processing is imposed to allow for 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 κ -S_c relationship can be used to derive the CCN spectrum from aerosol size-distribution to perform aerosol-CCN closure studies to further understand CCN impacts on warm clouds.

6.0 References

Kuang, C. 2016. Scanning Mobility Particle Spectrometer Instrument Handbook. U.S. Department of Energy, Atmospheric Radiation Measurement user facility, Richland, Washington. DOE/SC-ARM/TR-147.

Petters, MD, and SM Kreidenweis. 2007. "A Single Parameter Representation of Hygroscopic Growth and Cloud Condensation Nucleus Activity." *Atmospheric Chemistry and Physics* 7(8): 1961–1971, https://doi.org/10.5194/acp-7-1961-2007

Ren, J, F Zhang, Y Wang, D Collins, X Fan, X Jin, W Xu, Y Sun, M Cribb, and Z Li. 2018. "Using different assumptions of aerosol mixing state and chemical composition to predict CCN concentrations based on field measurements in urban Beijing." *Atmospheric Chemistry and Physics* 18(9): 6907–6921, https://doi.org/10.5194/acp-18-6907-2018

Uin, J. 2016. Ultra-High-Sensitivity Aerosol Spectrometer (UHSAS) Instrument Handbook. U.S. Department of Energy, Atmospheric Radiation Measurement user facility, Richland, Washington. DOE/SC-ARM/TR-163.

Uin, J. 2022. Cloud Condensation Nuclei Particle Counter Instrument Handbook. U.S. Department of Energy, Atmospheric Radiation Measurement user facility, Richland, Washington. DOE/SC-ARM/TR-168.

Appendix A

Quality Checks Applied in aosccn2cola.b1 Datastream

Figure 8 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 8 was generated using the ARM_nc_display Matlab function (courtesy of Connor Flynn).

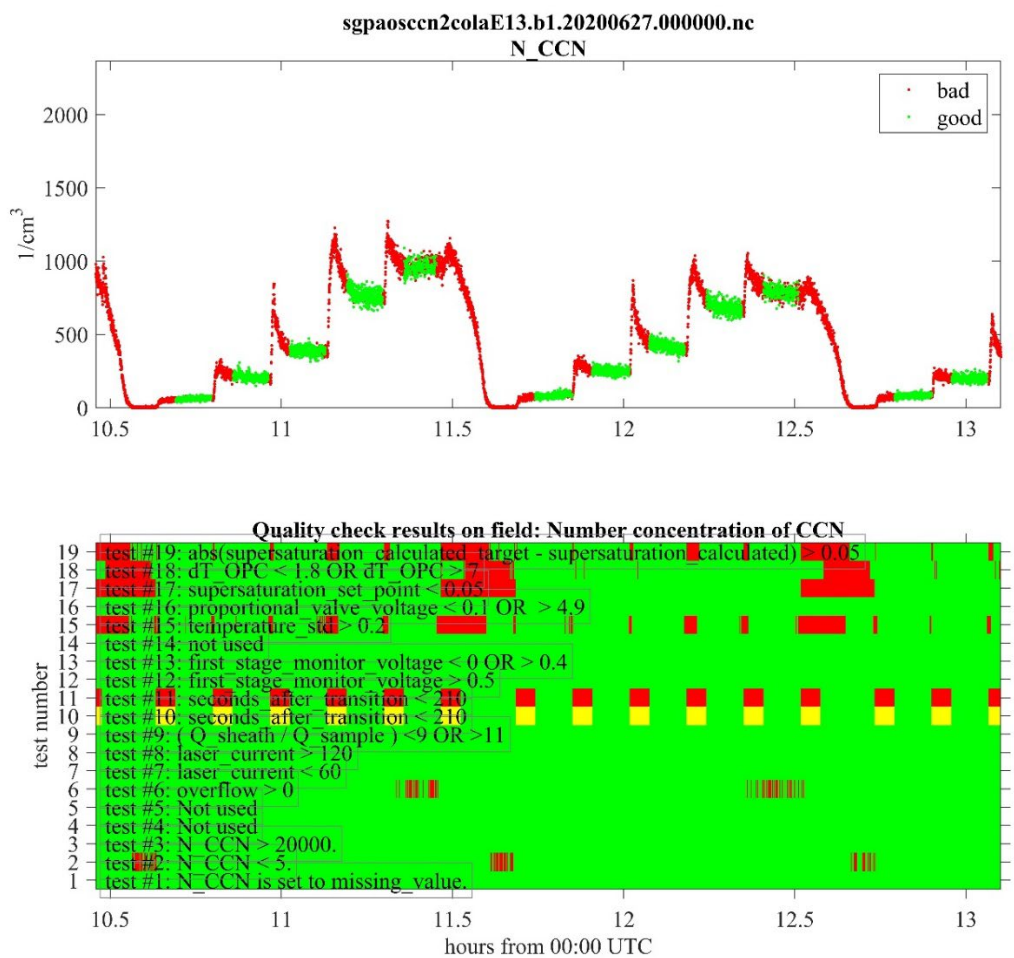

Figure 8. CCN concentrations and quality assessments from aosccn2cola.b1 at SGP E13.

ARM

www.arm.gov

U.S. DEPARTMENT OF ENERGY

Office of Science