Cloud Condensation Nuclei Closure During CLASIC 2007

Jason Tomlinson and Don Collins
PNNL
Texas A&M University
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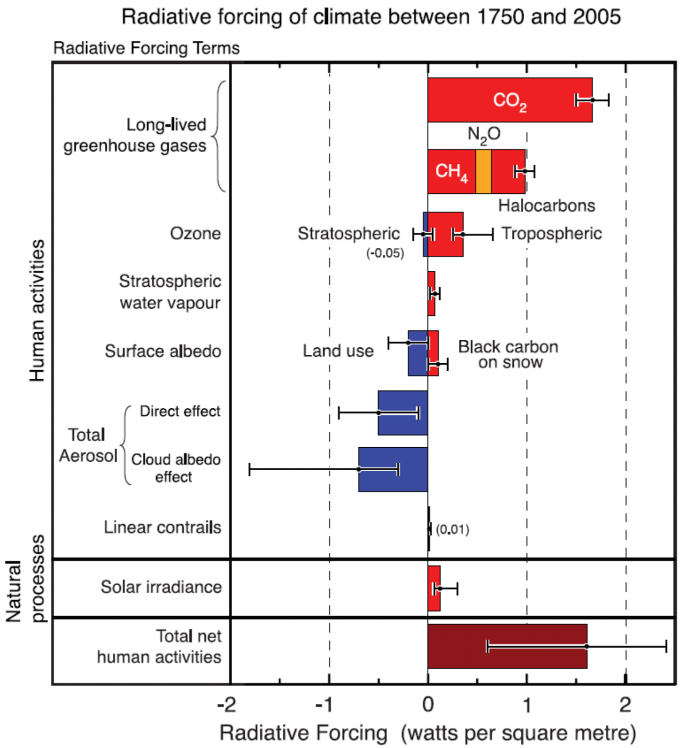
Introduction

- A major redesign of the Texas A&M Differential Mobility Analyzer/Tandem Differential Mobility Analyzer (DMA/TDMA) occurred in 2005/2006
  - Simultaneous measurements of size distributions and the hygroscopic properties of the aerosol
- For the CLASIC campaign, a Droplet Measurement Technologies (DMT) Cloud Condensation Nuclei Counter (CCNc) was also operated on the aircraft to measure the concentration of CCN active at supersaturations of 0.165, 0.305, and 0.60 %

Motivation

Introduction
Methodology
CLASIC 2007
Summary

- Characterize the aerosol size distributions and physicochemical properties in various environments
- Predict the effect on visibility and cloud condensation nuclei formation
  - Compare predicted versus measured CCN to ascertain our level of understanding particle activation
- Important to constrain results for global and regional climate models



IPCC 2007

DMA/TDMA

Introduction

Methodology

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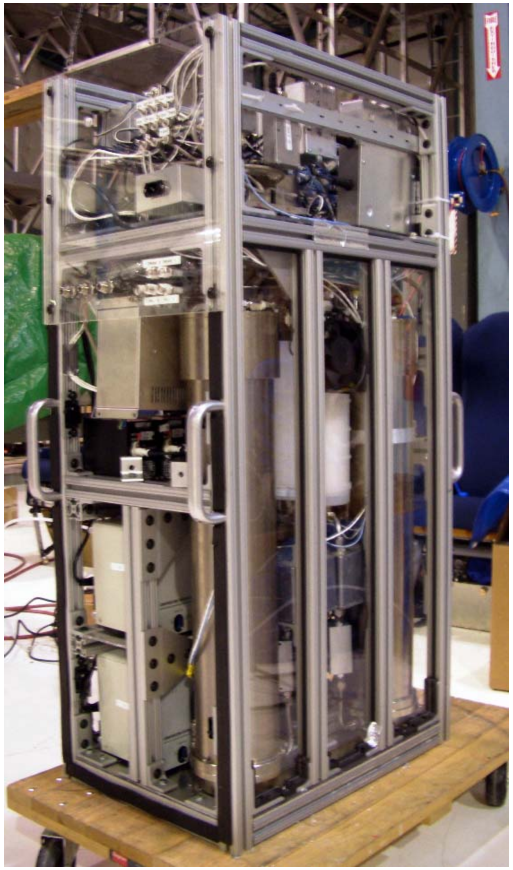
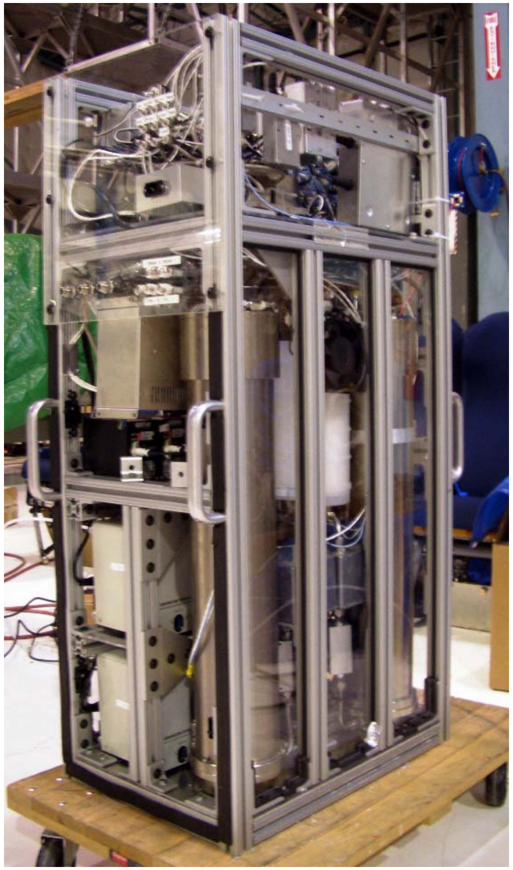
Summary



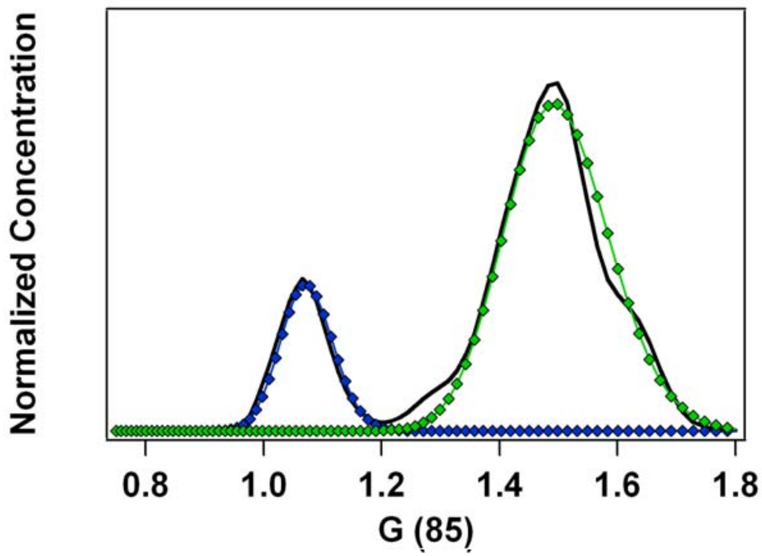
- 3 Hi-Flow Differential Mobility Analyzers
  - Operated at 10:1 flow ratio
- DMA size distribution measured from 0.010 to .700 μm every 90s
- TDMA growth distributions for dry diameters of 0.025, 0.050, 0.100, 0.200, and 0.400 μm measured every 12 minutes
- Maintained at 85% and 38^{0}C throughout the project

DMA/TDMA



- Calibrated at the beginning of CLASIC
- DMA
  - Inject PSL
- TDMA
  - Inject atomized ammonium sulfate and sodium chloride
  - Daily dry scans

DMA/TDMA



	Mode 1	Mode 2
N	0.24	0.76
GF	1.07	1.49
σ	1.04	1.06

- A log-normal distribution was fitted to each mode. The geometric means of the log-normals are assumed to represent the hygroscopicity of the aerosol population.

DMT CCNc

Lance et al., 2006

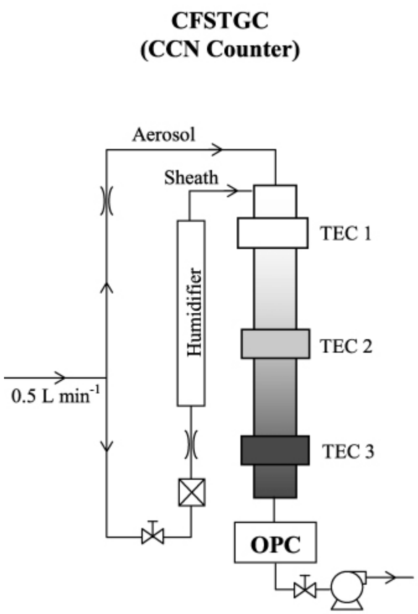
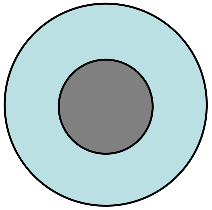


Image Source: DMT

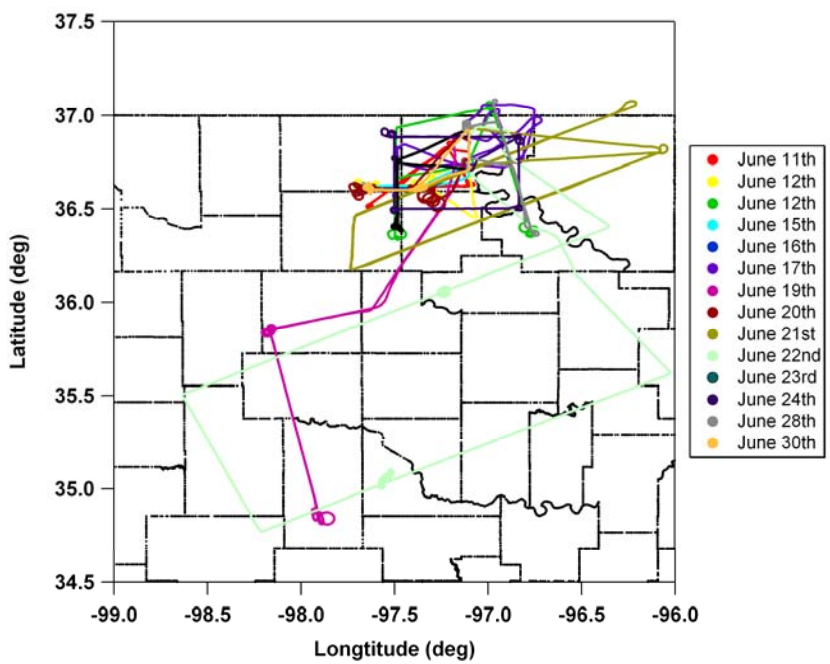
- Atomized ammonium sulfate is used to calibrate

CCN Prediction

- Use modified Köhler Theory to determine S_c value of a particle with known dry diameter and G(85)
- Assumptions:
  - Particle is composed of an insoluble core surrounded by a soluble shell
  - Soluble material is composed of Ammonium Sulfate with a van't Hoff factor (i) of 2.3
  - In subsaturated conditions, the vapor pressure over the particle is in equilibrium with ambient vapor pressure
  - Constant TDMA Temperature of 311 K



CLASIC 2007



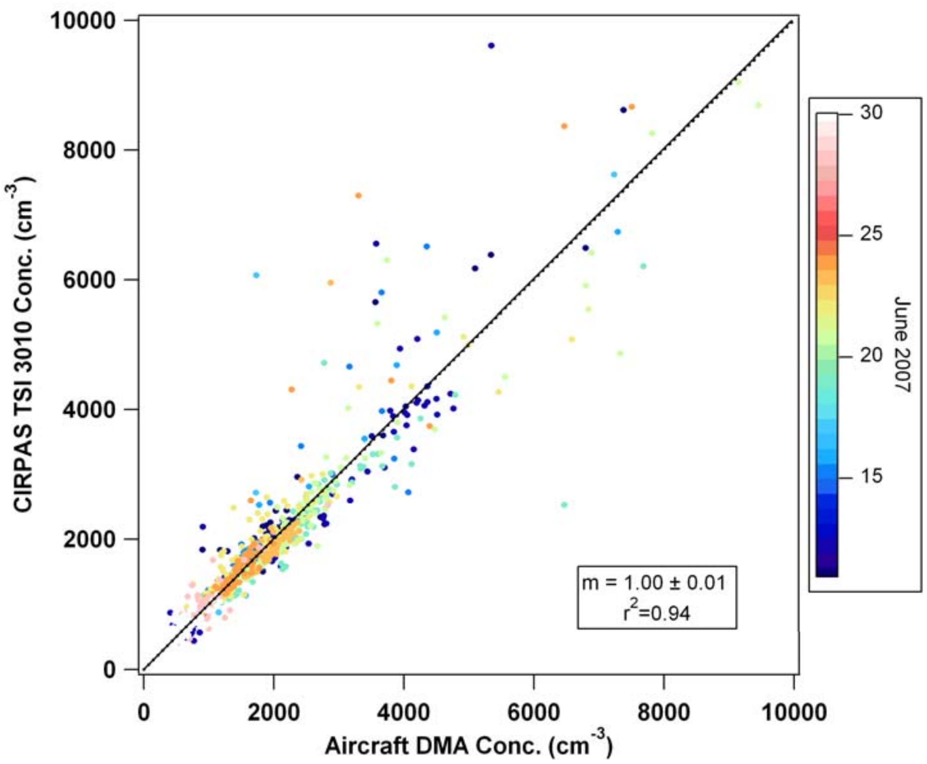
- 14 research flights were flown to study how land surface processes affect atmospheric aerosol loading and chemistry and the resulting effects on the microphysical and macrophysical properties of cumulus cloud fields
- Measured over 1300 size distributions
- 110 hygroscopic growth factor distributions for each dry diameter of 0.025, 0.050, 0.100, 0.200, and 0.400 μm
- DMT CCNc operated simultaneously

Pacific Northwest National Laboratory
Office of Science
U.S. DEPARTMENT OF ENERGY

CLASIC 2007

	June 2007													
	11	12	12	15	16	17	19	20	21	22	23	24	28	30
Size Distributions	■	■	■	■	■	■	■	■	■	■	■	■	■	■
G (85)	■	■	■	■	■	■	■	■	■	■	■	■		■
0.165%	■	■	■	■	■	■	■		■			■	■	
0.305%	■	■	■	■	■	■	■	■	■	■	■	■	■	■
0.600%	■	■	■	■	■	■	■		■			■	■	

Intercomparison



- Intercomparison between integrated number concentration and measured concentration from the CIRPAS TSI 3010

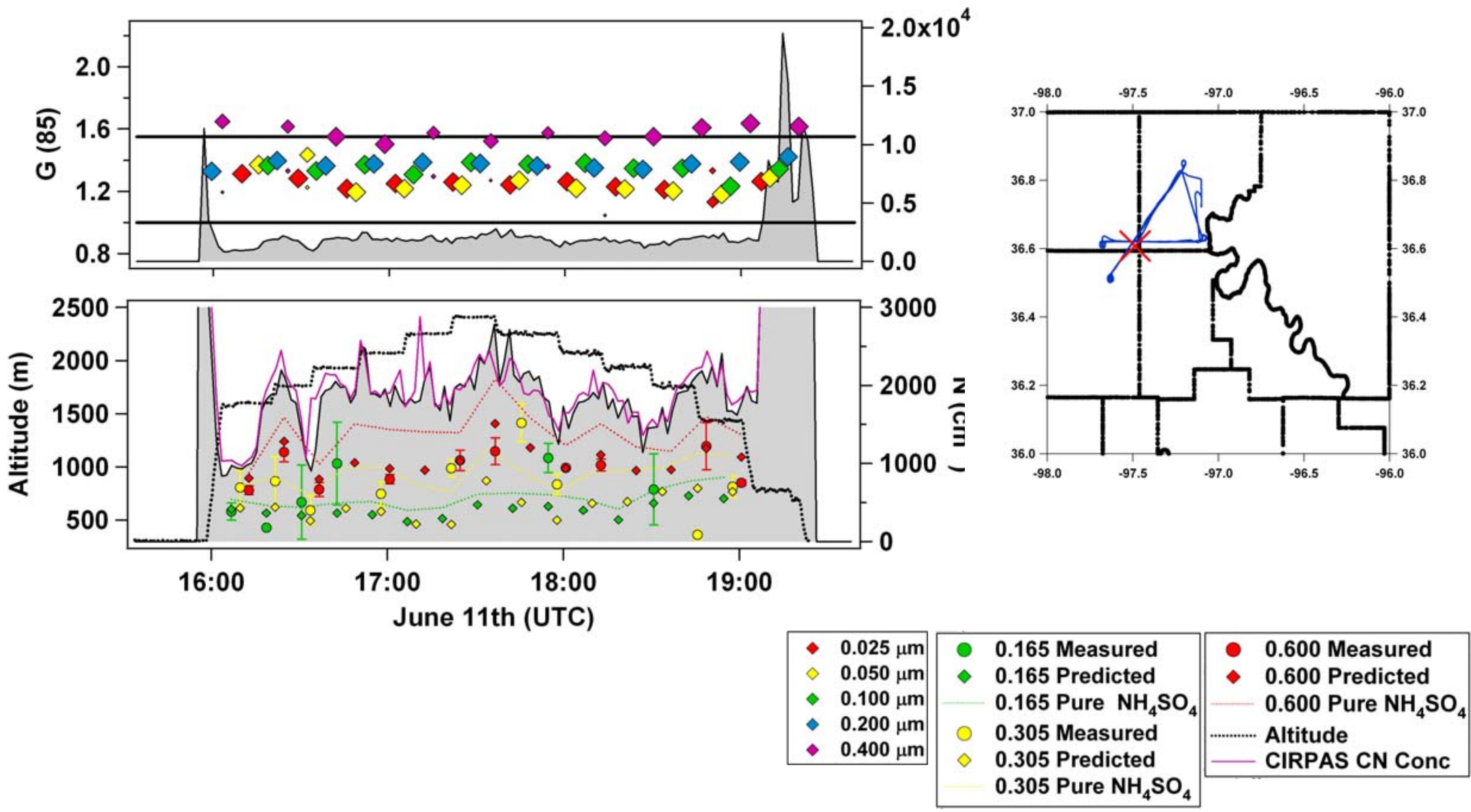
DMT CCNc

- For most flights the CCN counter was operated as follows:

Green	0.165 Supersaturation
Yellow	0.305 Supersaturation
Red	0.600 Supersaturation

- Research flights with rapid altitude changes the supersaturation was fixed at 0.305 %
- Measured CCN concentrations were passed through several quality control checks
  - Are the flows within 10% of flows during calibration?
  - Does the first bin contribute a significant fraction to the overall counts?
  - Are the measured values greater than one would expect for pure ammonium sulfate?
- Number of data points after QC
  - 0.165 %: n=22 0.305 %: n=37
    Fixed 0.305%: n=33 0.600 %: n=49

June 11th Time Series



June 19th Time Series

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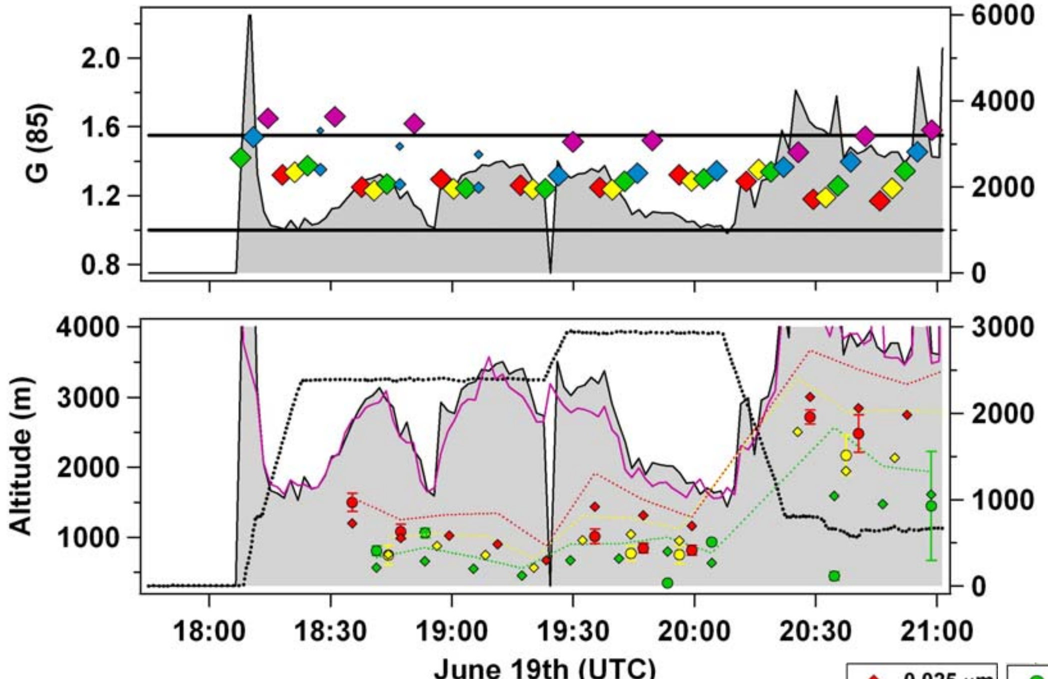
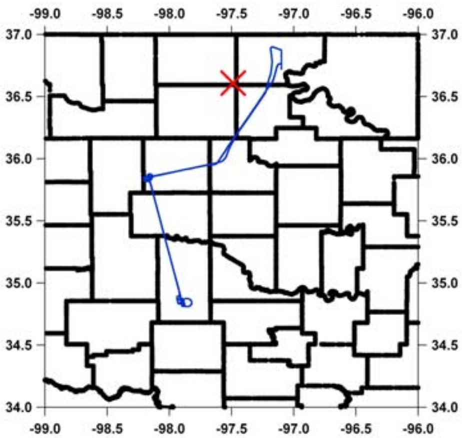
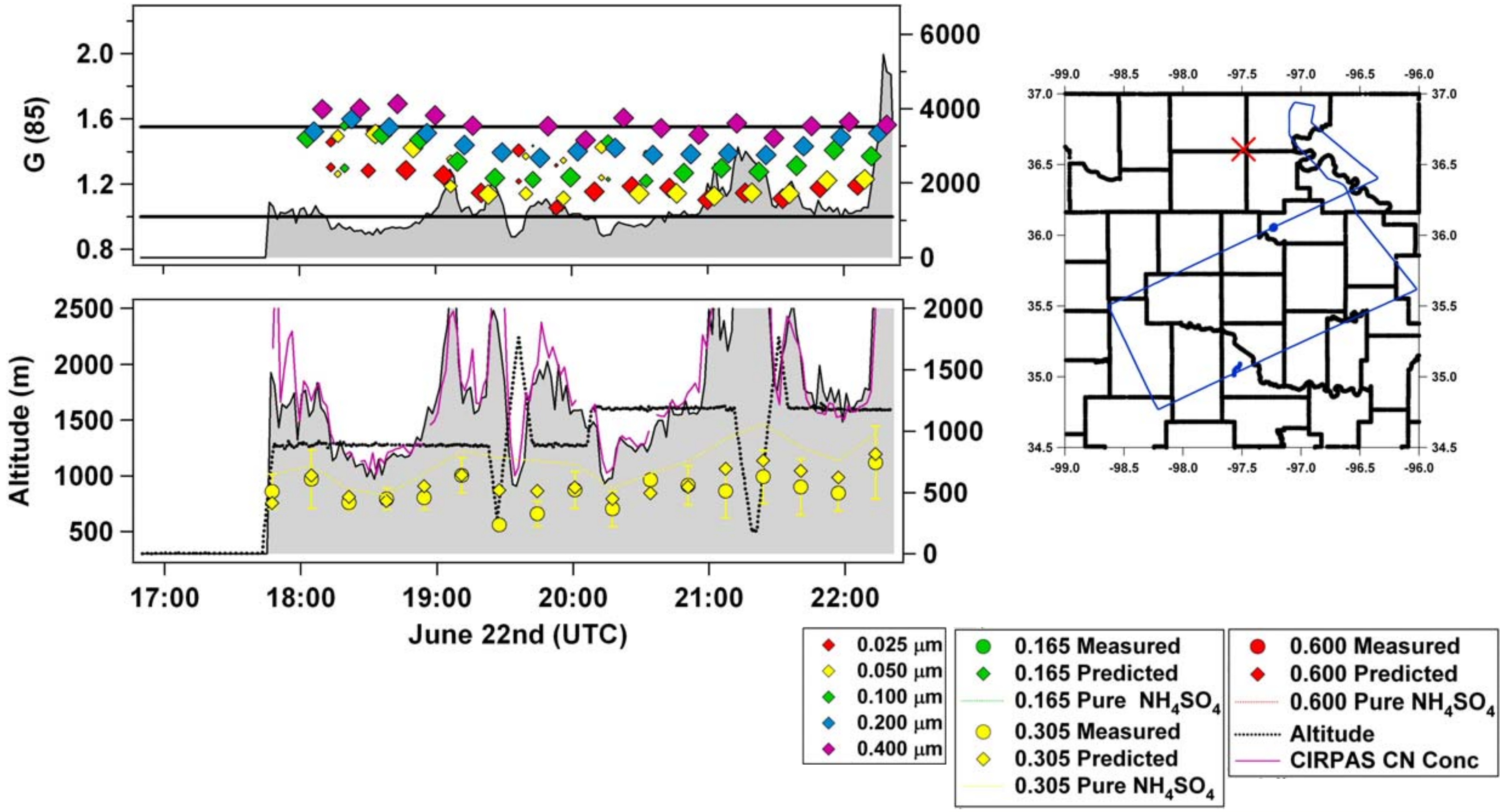




Web Page

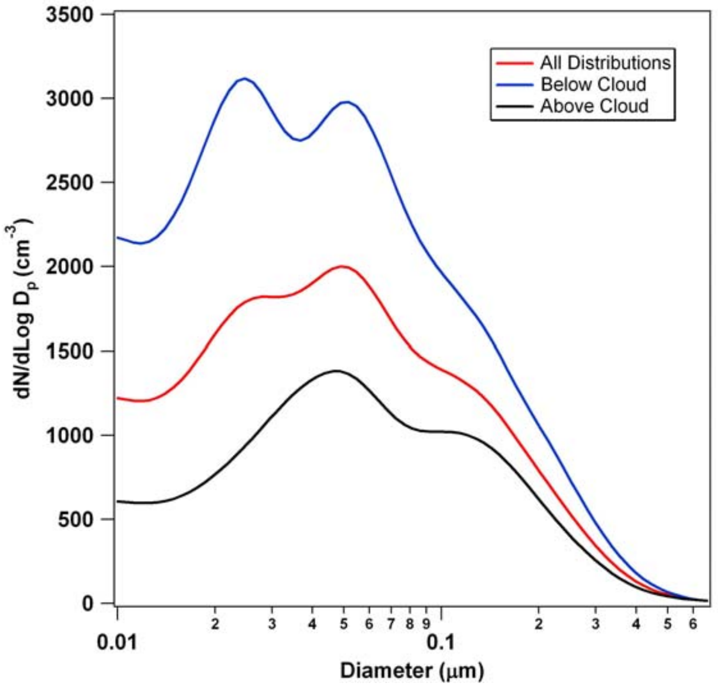
June 22nd Time Series



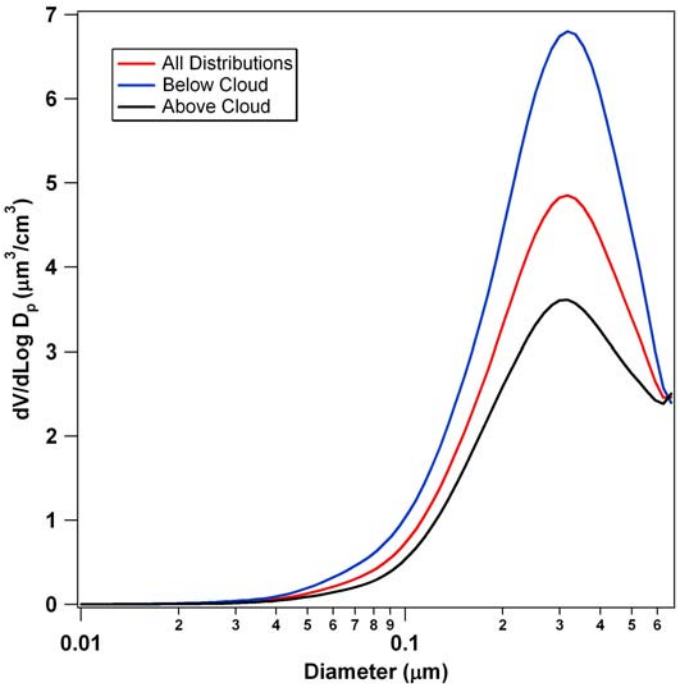
Web Page

Size Distributions

Number Distribution



Volume Distribution



G (85)

Introduction

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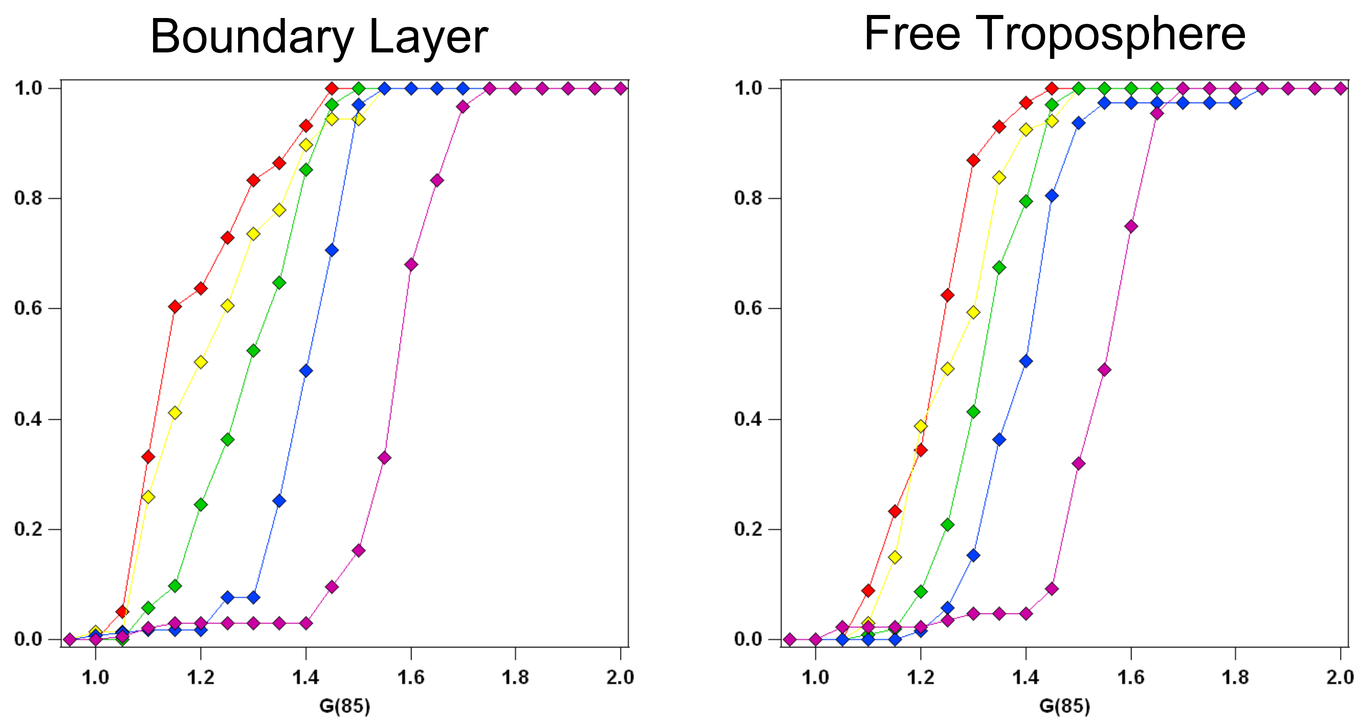
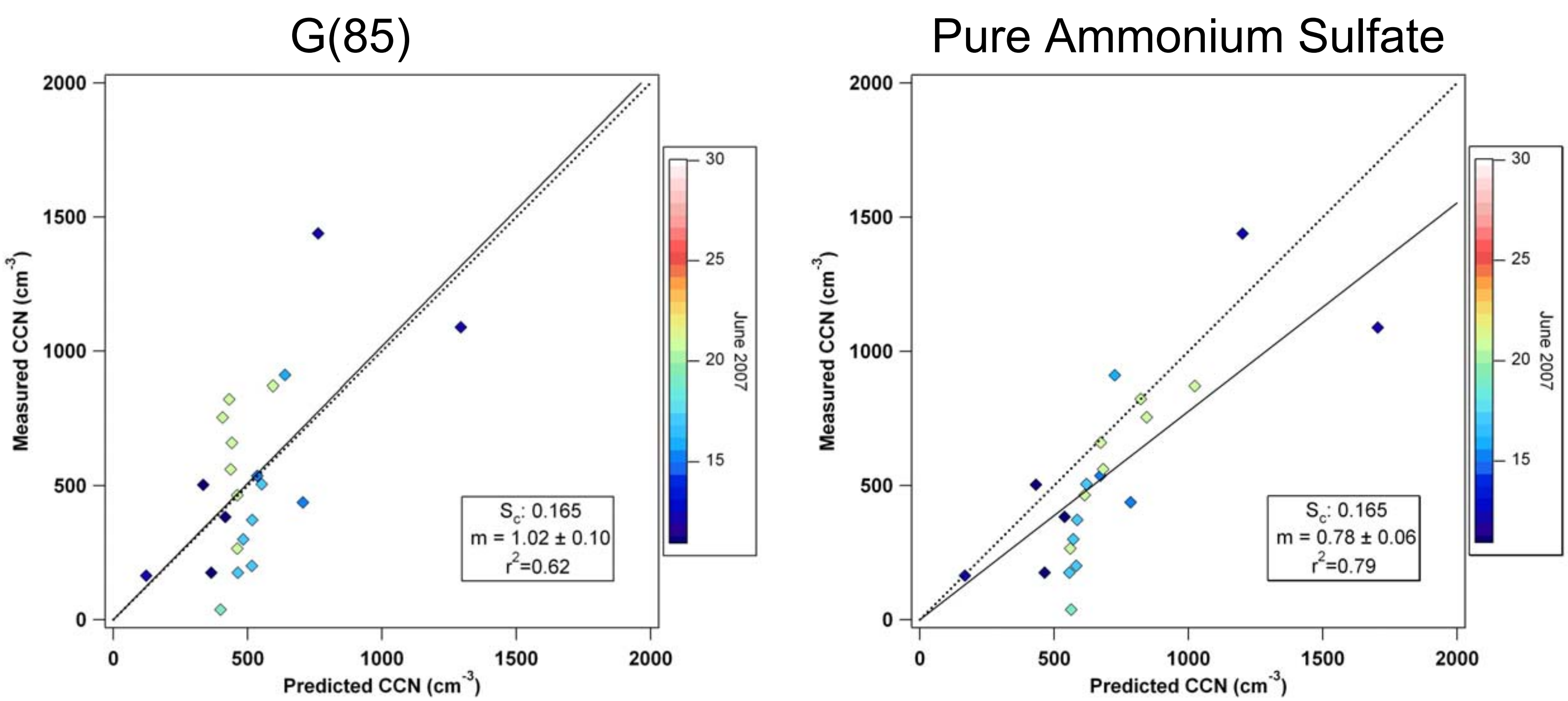
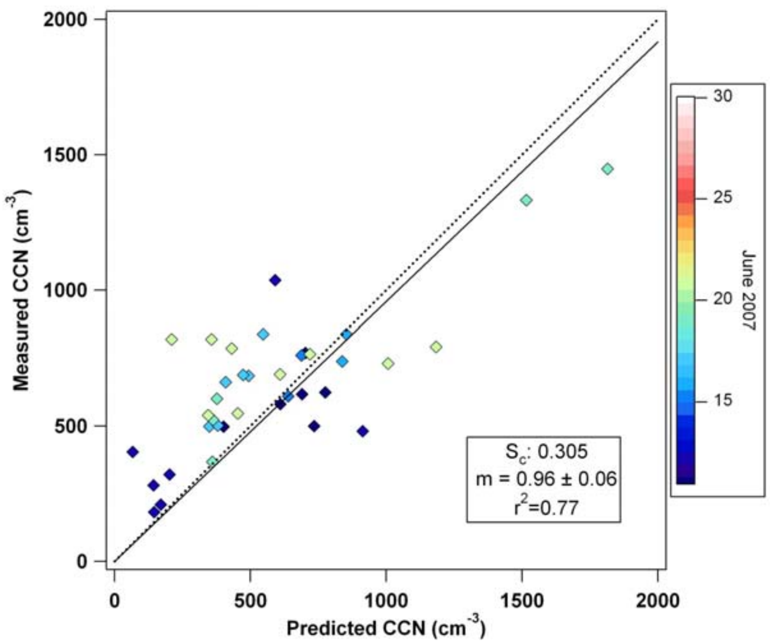


CCN Scatterplot (0.165)

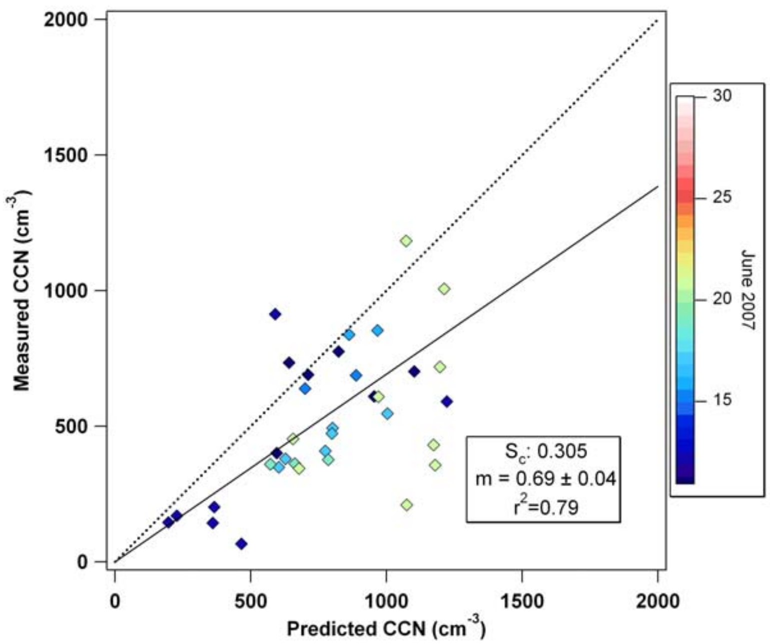


CCN Scatterplot (0.305)

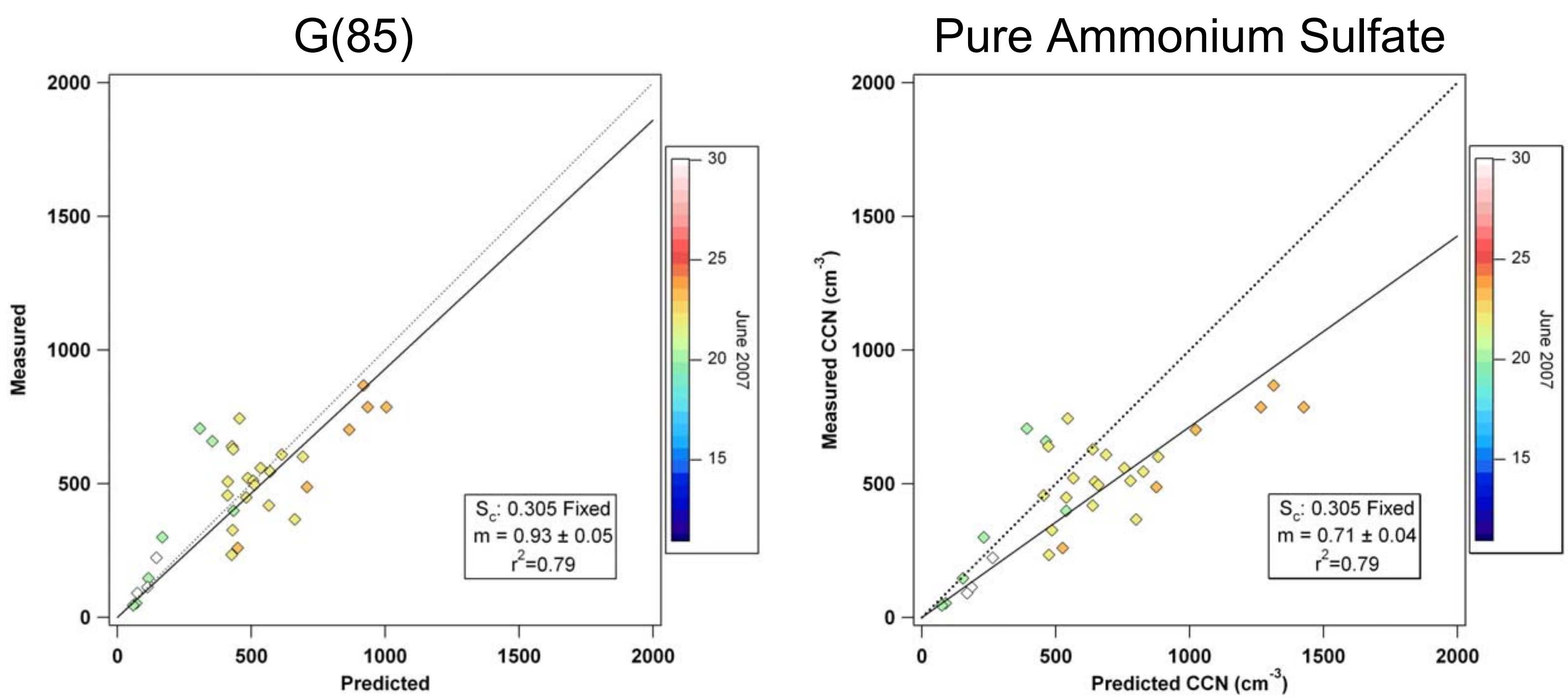
G(85)



Pure Ammonium Sulfate

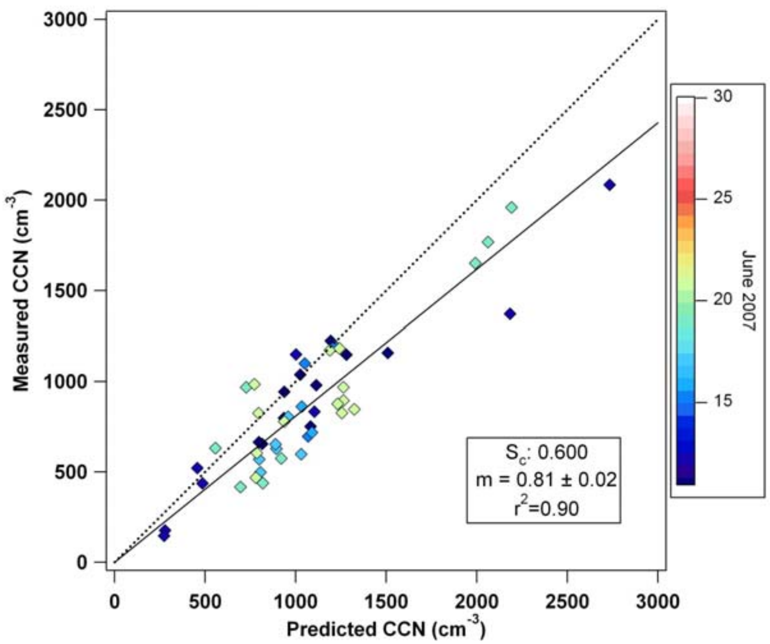


CCN Scatterplot (0.305 Fixed)

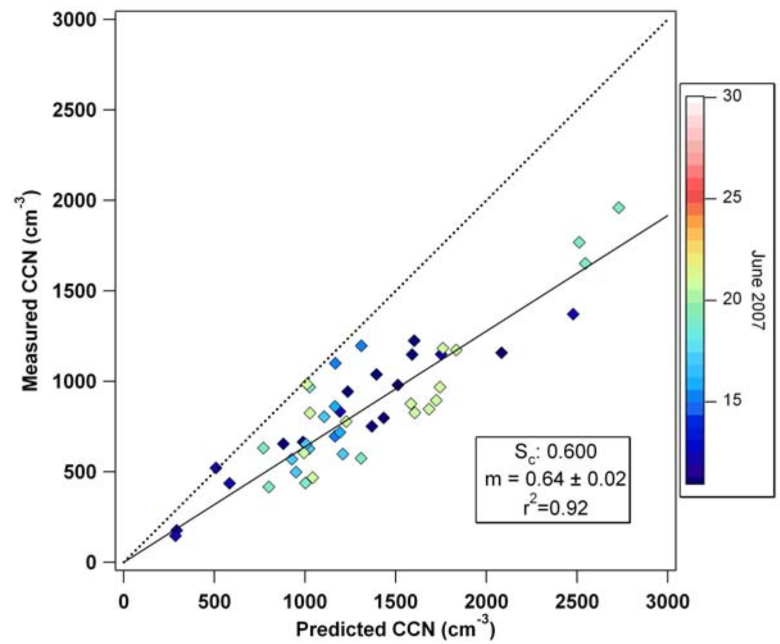


CCN Scatterplot (0.60)

G(85)



Pure Ammonium Sulfate



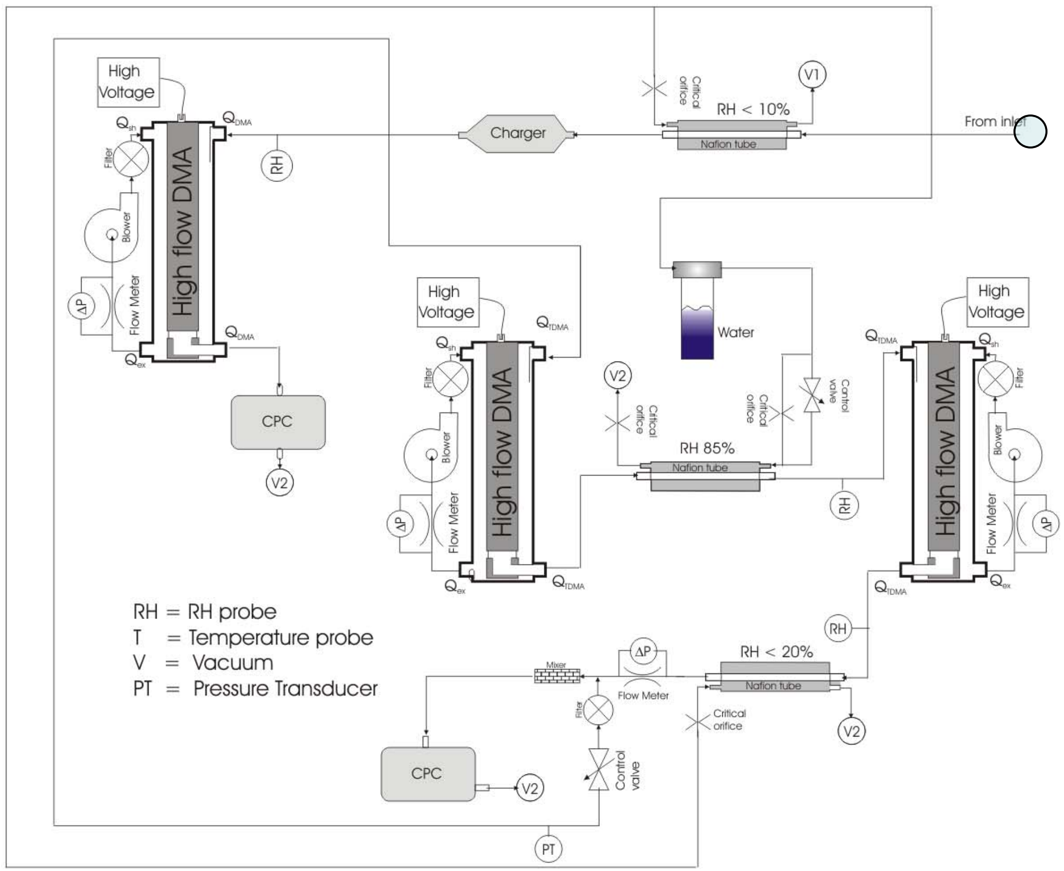
Summary

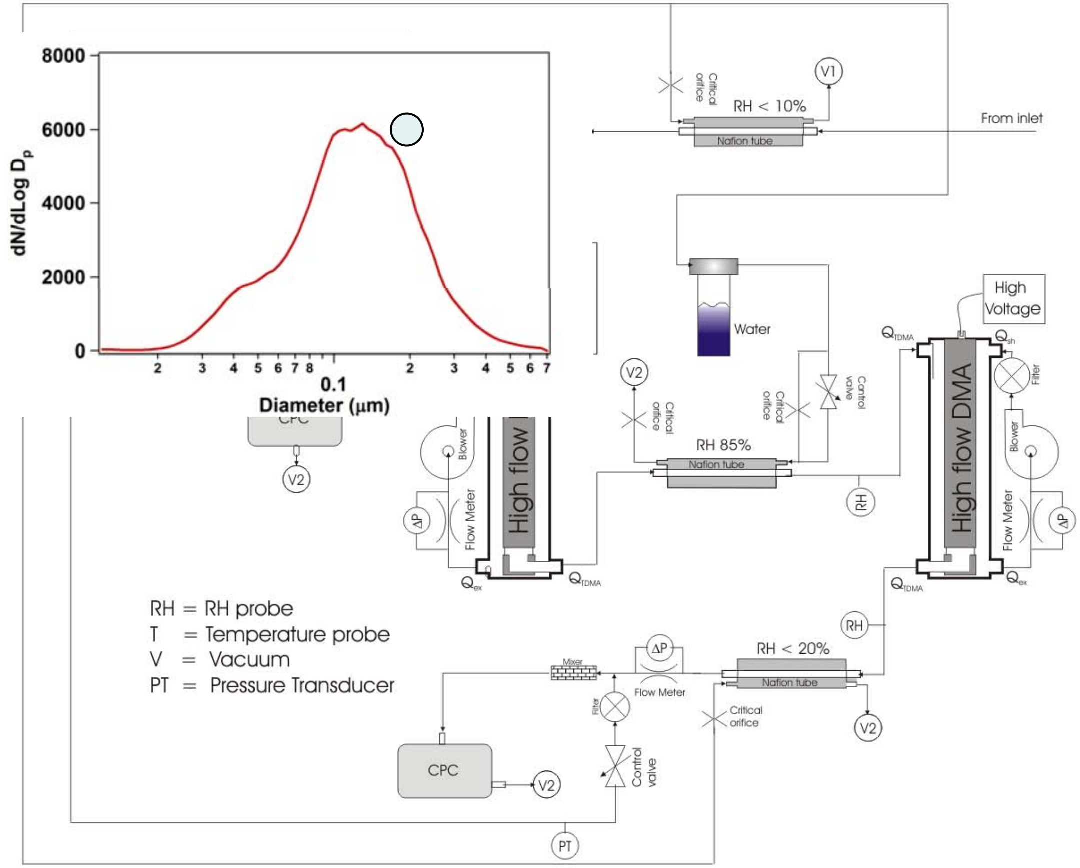
- During the CLASIC 2007 campaign a DMA/TDMA and DMT CCNc were operated over 14 research flights measuring size distribution, hygroscopic growth distributions, and the number of CCN that would activate at 0.165 %, 0.305%, and 0.600%
- The average size distribution indicate a very stable volume mode and larger concentration within the boundary layer
- The hygroscopic growth distributions indicate the presence of organics
- Predicted CCN concentration agree well with measured CCN concentrations at supersaturations of 0.165% and 0.305%. However over prediction occurs at a supersaturation of 0.600%

Thank You

http://www.met.tamu.edu/research/aerosol/Webpage/CLASIC_pages/CLASICcalendar607.html

Extra Slides


High Voltage
High flow DMA
Q_sh
Q_DMA
Q_ex
Filter
Blower
Flow Meter
ΔP
RH
CPC
V2
Charger
Critical orifice
RH < 10%
Nafion tube
V1
From inlet
Water
Control valve
RH 85%
Q_TDMA
RH < 20%
Mixer
PT
RH = RH probe
T = Temperature probe
V = Vacuum
PT = Pressure Transducer



8000
6000
4000
2000
0
dN/dLog D_p
2 3 4 5 6 7 8
0.1
2 3 4 5 6 7
Diameter (μm)
Critical orifice
V1
RH < 10%
From inlet
Nafion tube
Water
High Voltage
Q_TDMA
Q_sh
V2
Critical orifice
Control valve
Critical orifice
Filter
RH 85%
Nafion tube
RH
Blower
High flow DMA
Blower
High flow
CPC
V2
Flow Meter
ΔP
Flow Meter
ΔP
Q_ex
Q_TDMA
Q_TDMA
Q_ex
RH
RH = RH probe
T = Temperature probe
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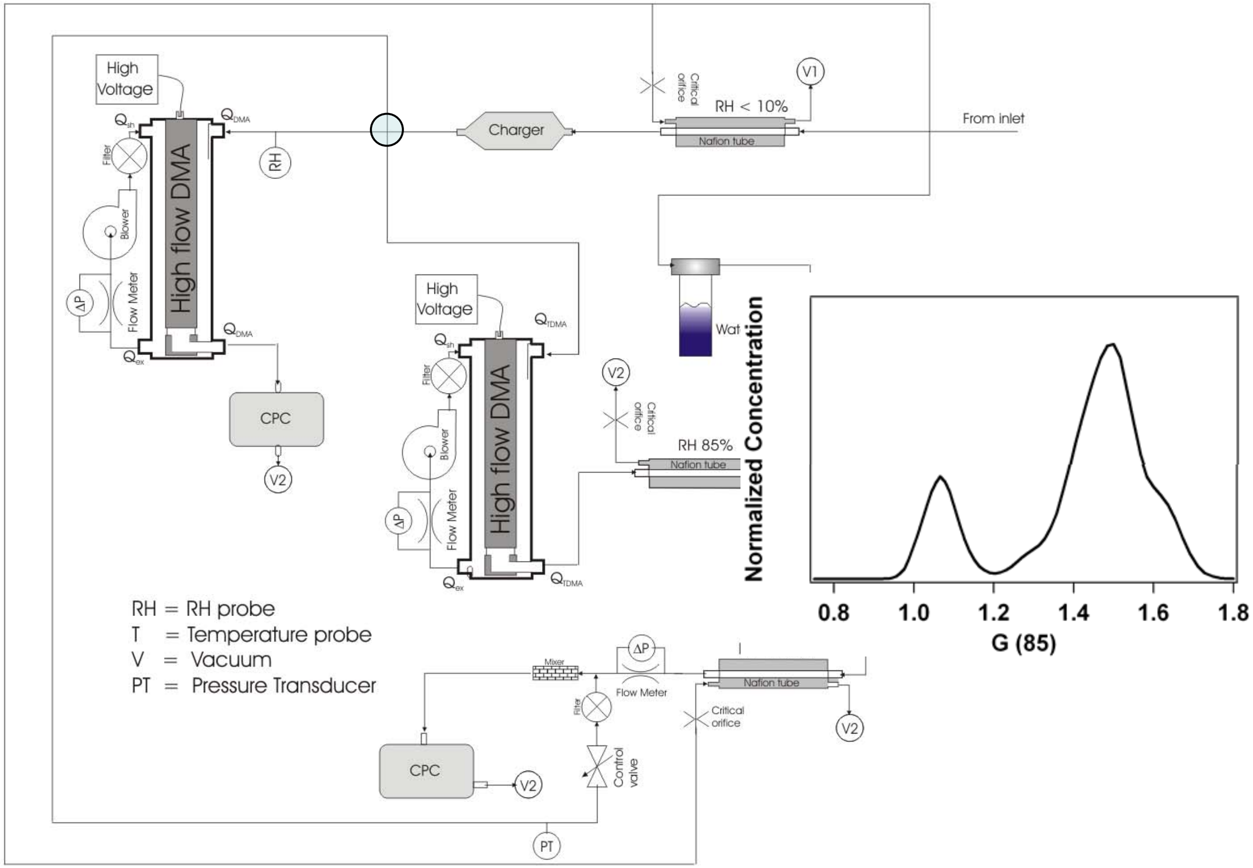
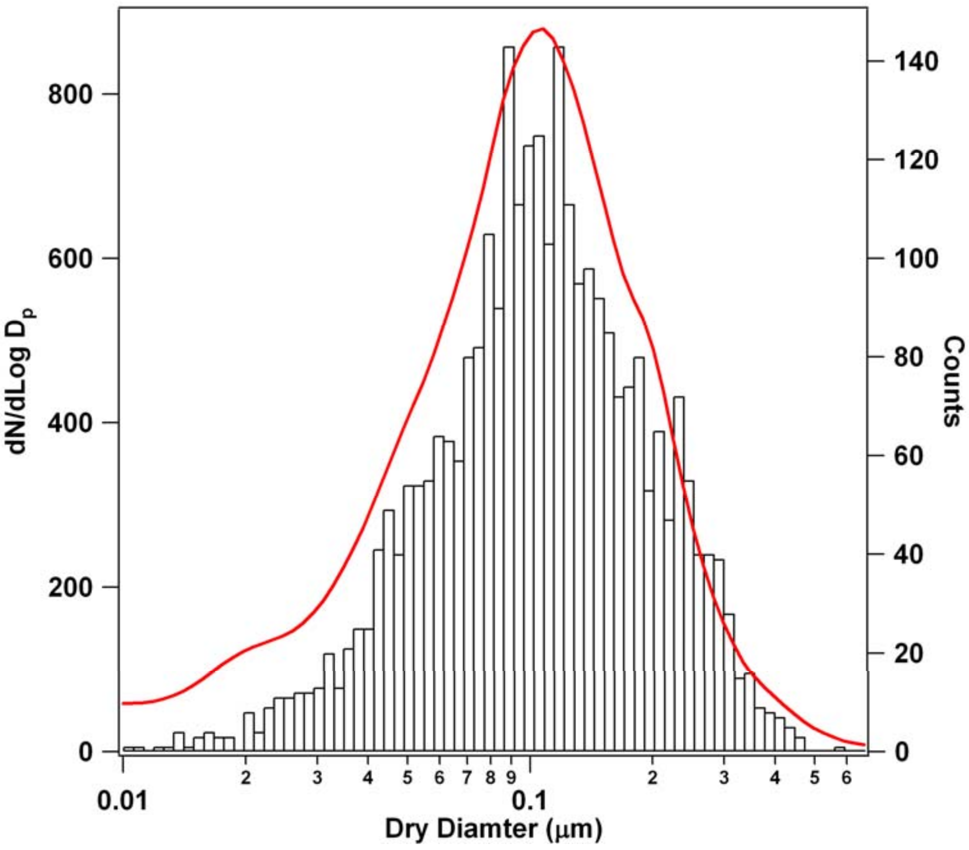
PT = Pressure Transducer
Mixer
ΔP
RH < 20%
Flow Meter
Nafion tube
Filter
Critical orifice
V2
CPC
V2
Control valve
PT



High Voltage
High flow DMA
Charger
RH < 10%
Nafion tube
From inlet
V1
RH
CPC
V2
Flow Meter
Blower
Filter
Critical orifice
RH 85%
Wat
Control valve
PT
Mixer
RH = RH probe
T = Temperature probe
V = Vacuum
PT = Pressure Transducer
Normalized Concentration
0.8
1.0
1.2
1.4
1.6
1.8
G (85)

- The size distributions for the DMA are recovered with a Twomey Algorithm

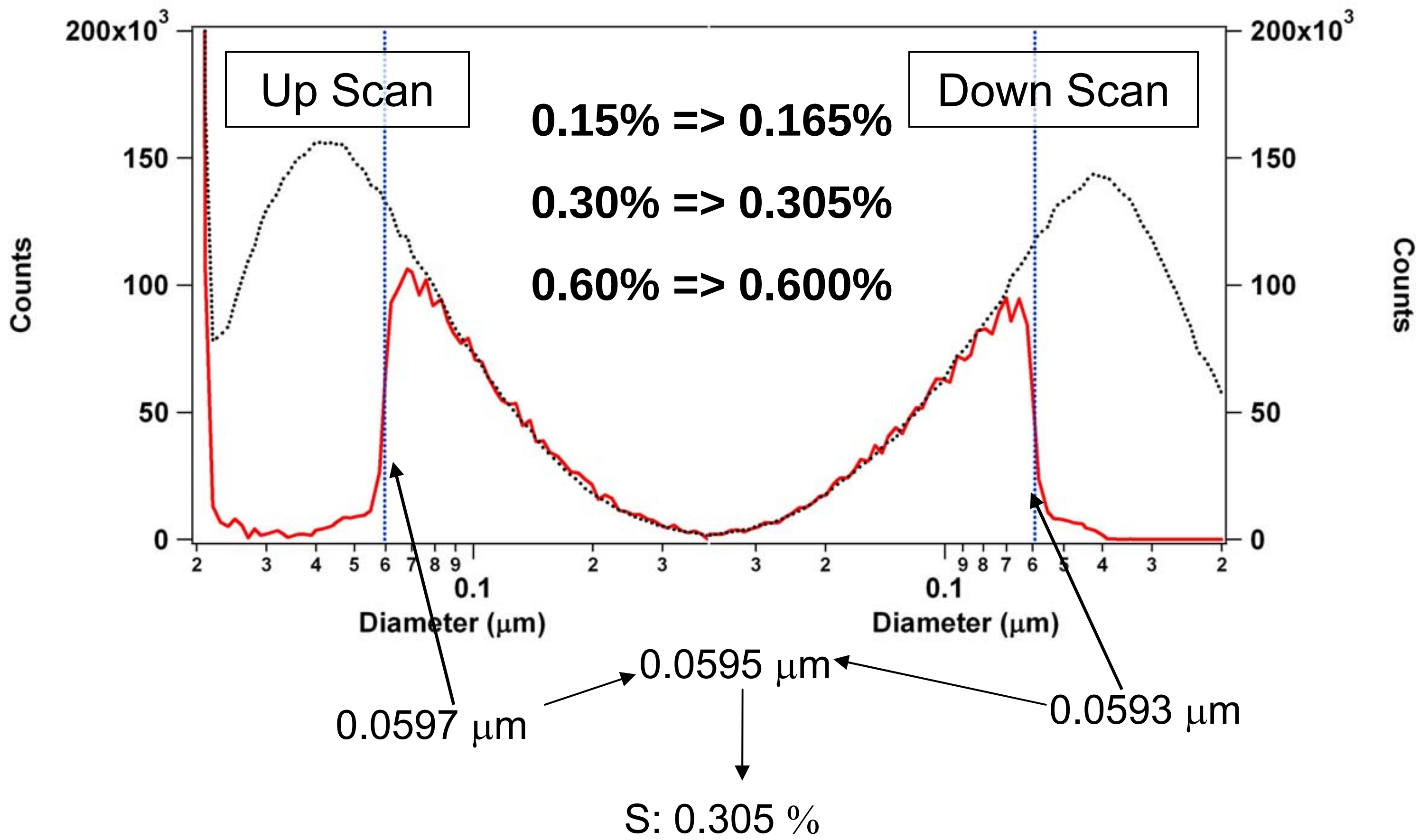


Integrated Concentration
Number: 591.4 /cm^3
Surface: 36.8 µm2/cm3
Volume: 1.5 µm3/cm3

CCNc Calibration

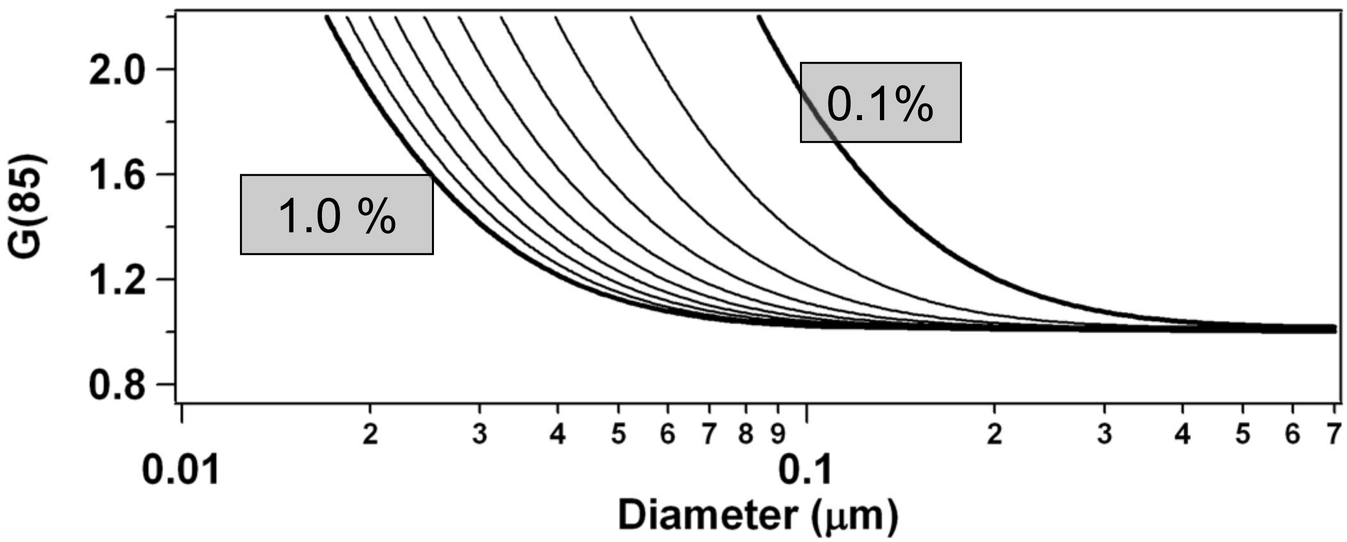
- Atomized ammonium sulfate is passed through a DMA
  - 0.020 up to 0.400 μm
  - 15 minute scan time
- 50% point is determined to be the activation diameter
- Kohler theory is used to calculate the corresponding supersaturation

CCNc Calibration



CCNc Calibration

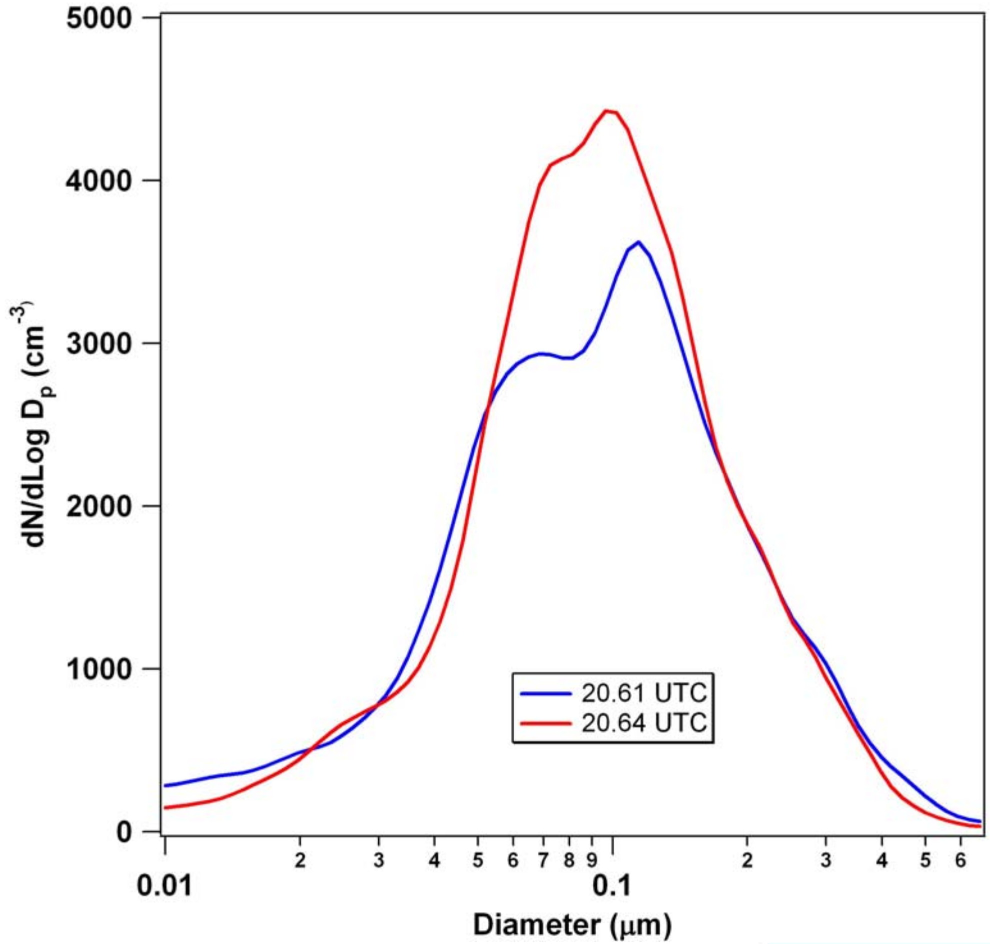
One decadal range of Sc values for Ammonium Sulfate



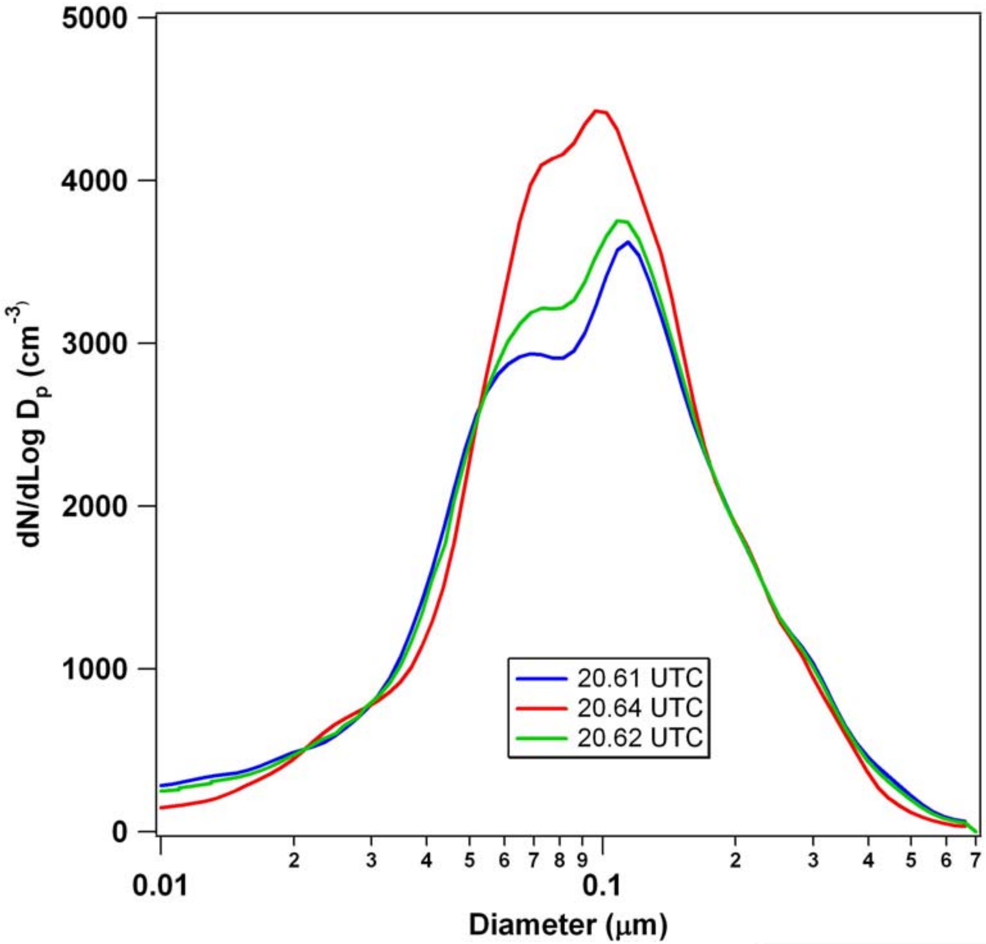
CCNc Calibration

- Example from June 19th at 20:37 UTC
- DMT CCNc measured roughly 1353 +/- 230 cm^{-3} activated at a supersaturation of 0.305%

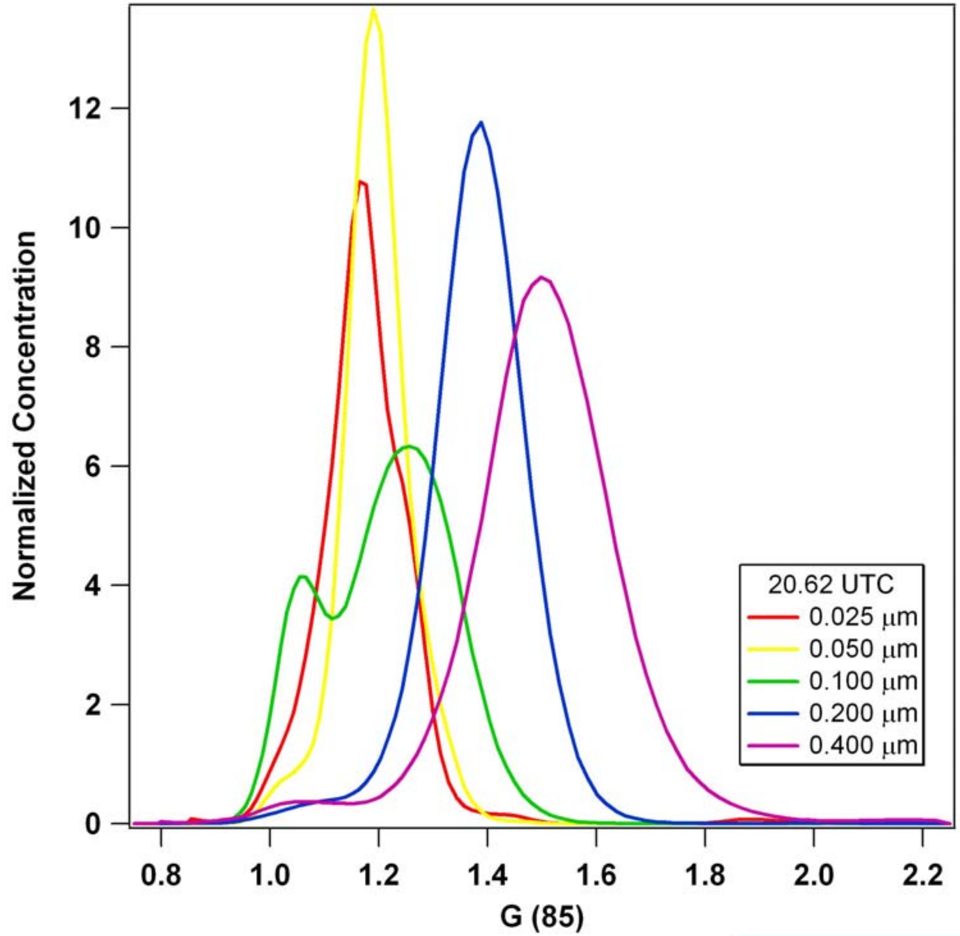
CCNc Calibration



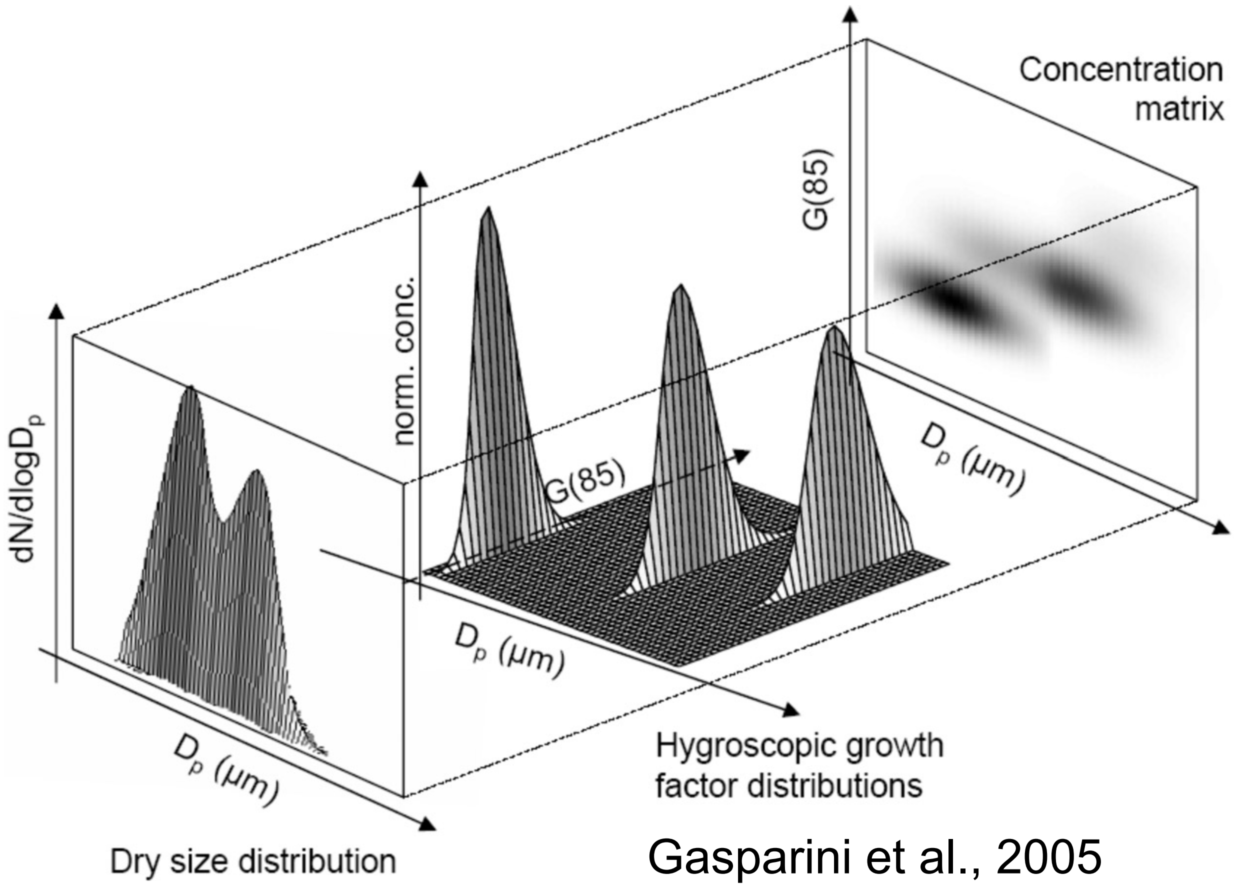
CCNc Calibration



CCNc Calibration

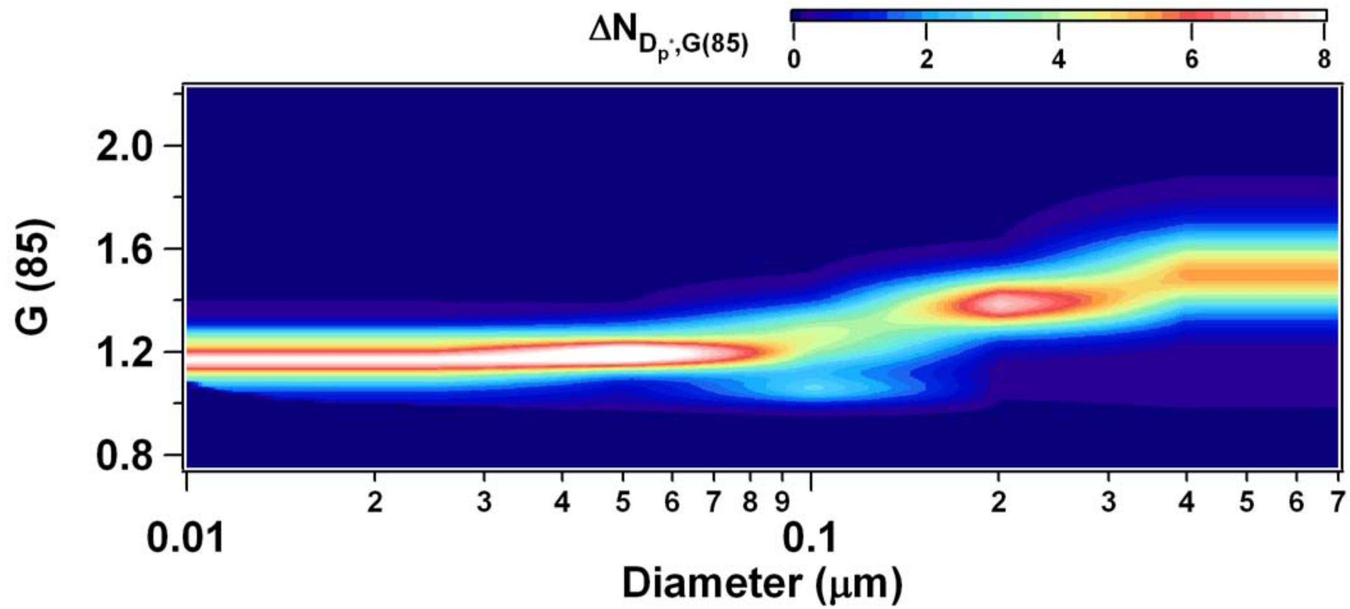


CCNc Calibration



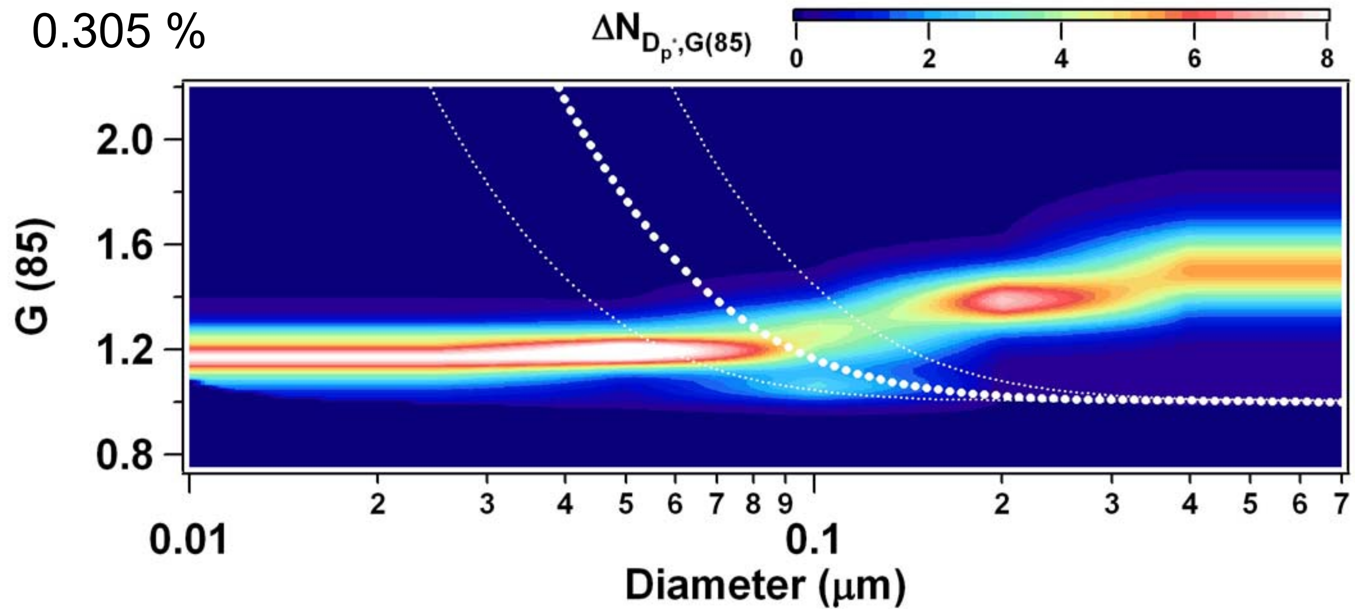
Gasparini et al., 2005

CCNc Calibration



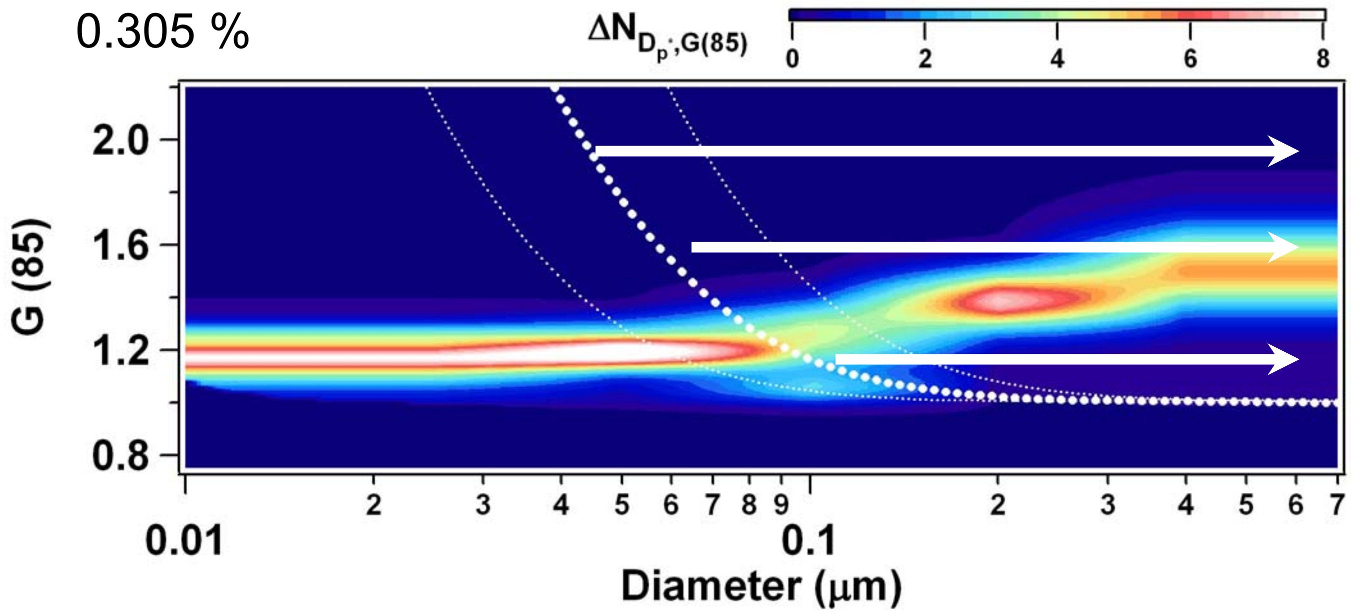
CCNc Calibration

0.305 %



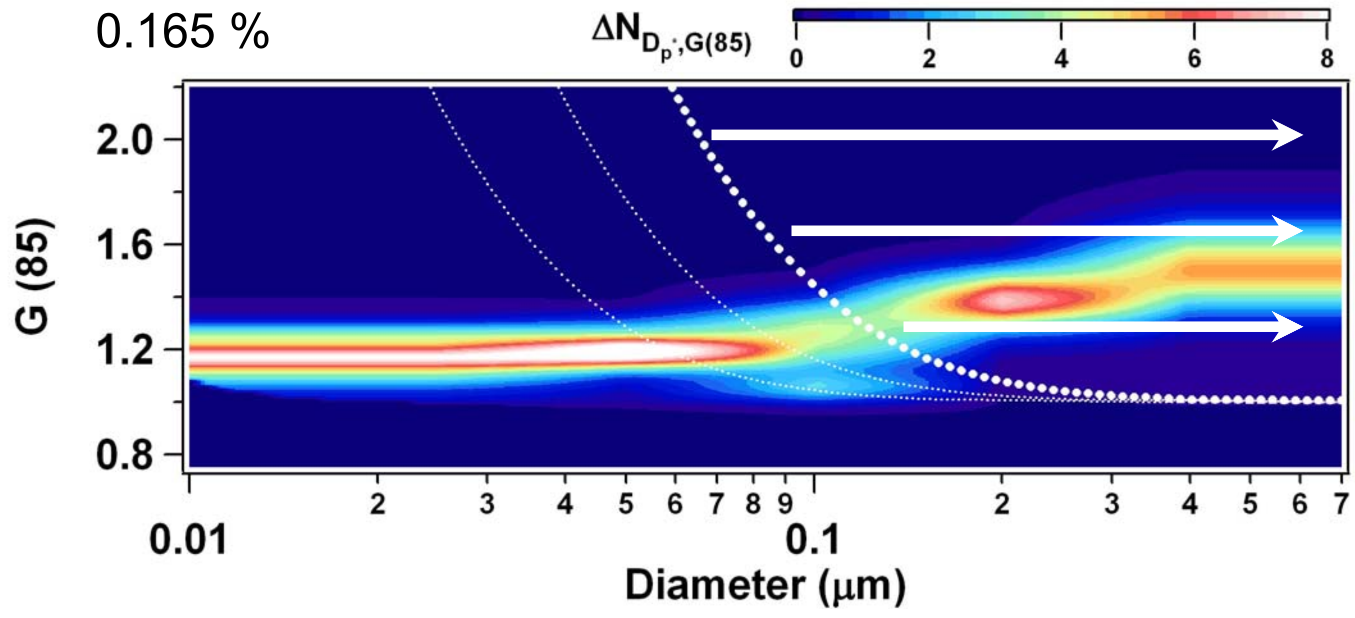
CCNc Calibration

0.305 %

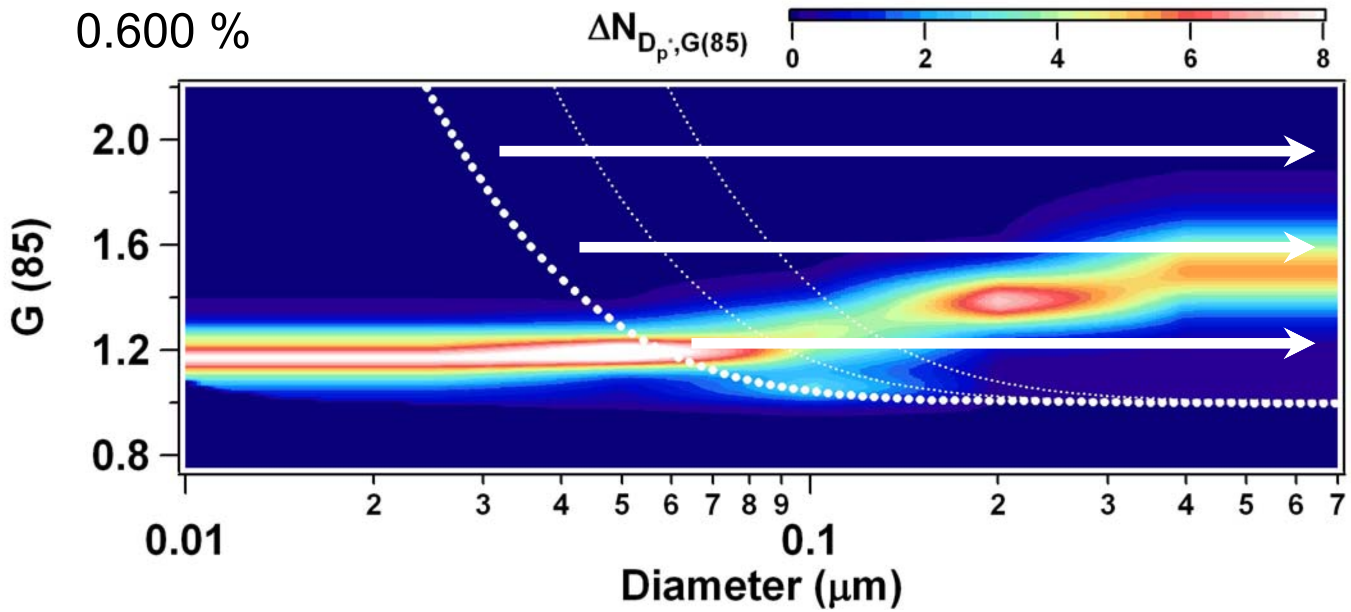


Predicted CCN Concentration: 1333 cm^{-3}

CCNc Calibration



CCNc Calibration



Growth Factor Prediction

Köhler Equation → $$0.85 = \alpha_w \exp\left[\frac{4M_w\sigma_{as}}{RT\rho_w(G(85)*D_p^*)}\right]$$

Growth Factor Prediction

Köhler Equation →

$$0.85 = \alpha_w \exp\left[\frac{4M_w\sigma_{as}}{RT\rho_w(G(85) * D_p^*)}\right]$$

$$V_{85} = \frac{\pi}{6} D_{p(85)}^3 = V_i + V_{as}$$

$$V_{dry} = \frac{\pi}{6} D_p^{*3} = V_i + V_s$$

→

$$\frac{m_s}{\varepsilon_s \rho_{as}} - \frac{m_s}{\rho_s} = \frac{\pi}{6}\left(D_{p(85)}^3 - D_p^{*3}\right)$$

Growth Factor Prediction

Köhler Equation → $$0.85 = \alpha_w \exp\left[\frac{4M_w\sigma_{as}}{RT\rho_w(G(85)*D_p^*)}\right]$$

$$V_{85} = \frac{\pi}{6}D_{p(85)}^3 = V_i + V_{as}$$

$$V_{dry} = \frac{\pi}{6}D_p^{*3} = V_i + V_s$$

→ $$\frac{m_s}{\varepsilon_s\rho_{as}} - \frac{m_s}{\rho_s} = \frac{\pi}{6}\left(D_{p(85)}^3 - D_p^{*3}\right)$$

Surface Tension → $$\sigma_{as} = \sigma_w(T) + \frac{2\times10^{-6}\varepsilon_s\rho_{as}}{M_s}$$

Growth Factor Prediction

Köhler Equation → $0.85 = \alpha_w \exp\left[\frac{4M_w\sigma_{as}}{RT\rho_w(G(85)*D_p^*)}\right]$

$$V_{85} = \frac{\pi}{6}D_{p(85)}^3 = V_i + V_{as}$$

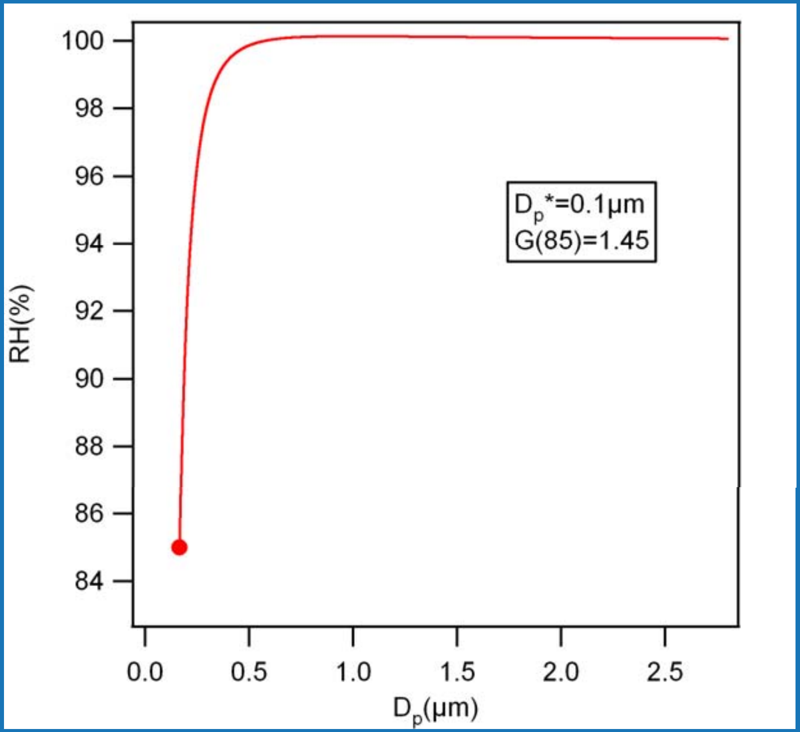
$$V_{dry} = \frac{\pi}{6}D_p^{*3} = V_i + V_s$$

→ $$\frac{m_s}{\varepsilon_s\rho_{as}} - \frac{m_s}{\rho_s} = \frac{\pi}{6}\left(D_{p(85)}^3 - D_p^{*3}\right)$$

Surface Tension → $\sigma_{as} = \sigma_w(T) + \frac{2\times10^{-6}\varepsilon_s\rho_{as}}{M_s}$

α_w and ρ_{as} from Tang and Munklewitz, 1994

Growth Factor Prediction



TDMA
Köhler Equation

→

Mass of
Solute

Growth Factor Prediction

Köhler Equation #2 ⟶ $$\frac{e'}{e_s} = \gamma_w \chi_w \exp\left[\frac{4 M_w \sigma_{as}}{RT \rho_w D_d}\right]$$

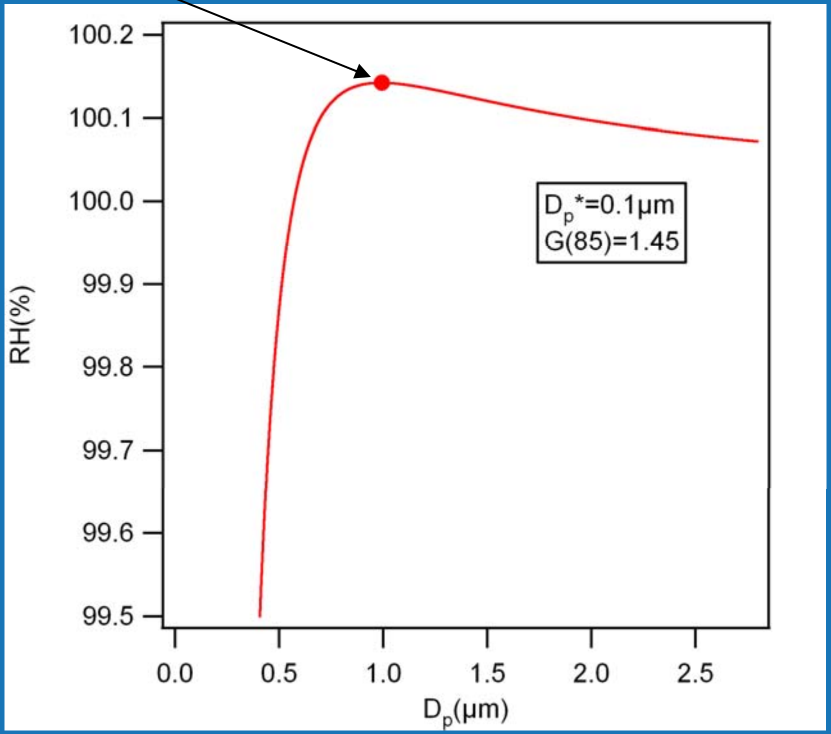
- The soluble mass determined in the initial calculations is fixed in the mass fraction of solute in solution term when calculating e'/e_s.
- Since this second calculation is representative of a particle in supersaturated conditions, we assume ideal behavior:

$$\gamma_w = 1 \qquad \chi_w = \frac{n_w}{n_w + i n_s} = \frac{1}{\frac{iM_w}{M_s}\left(\frac{\varepsilon_s}{1-\varepsilon_s}\right) + 1}$$

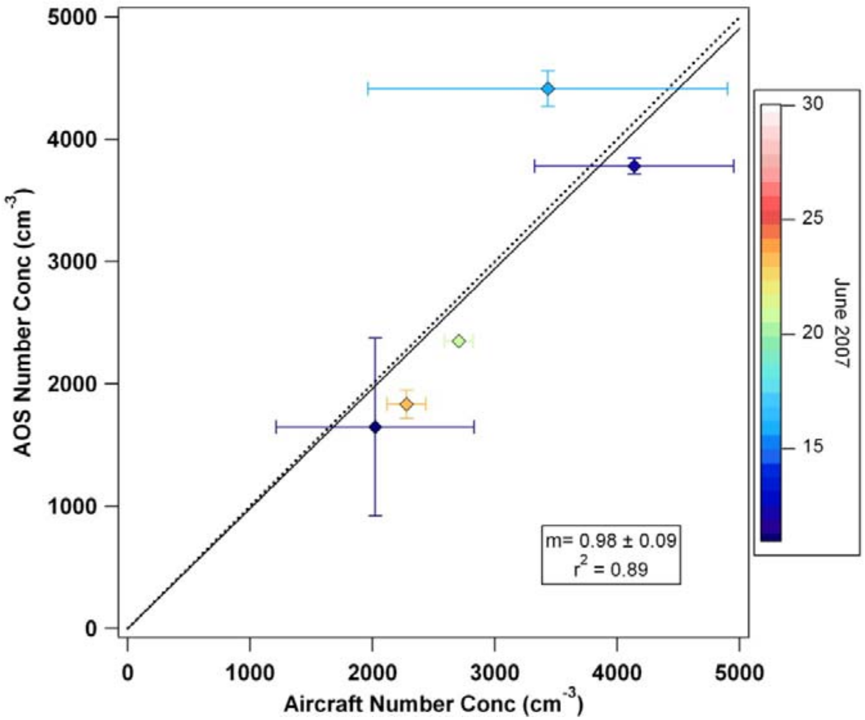
TDMA Köhler Equation ⟶ Mass of Solute ⟶ Köhler equation #2

Growth Factor Prediction

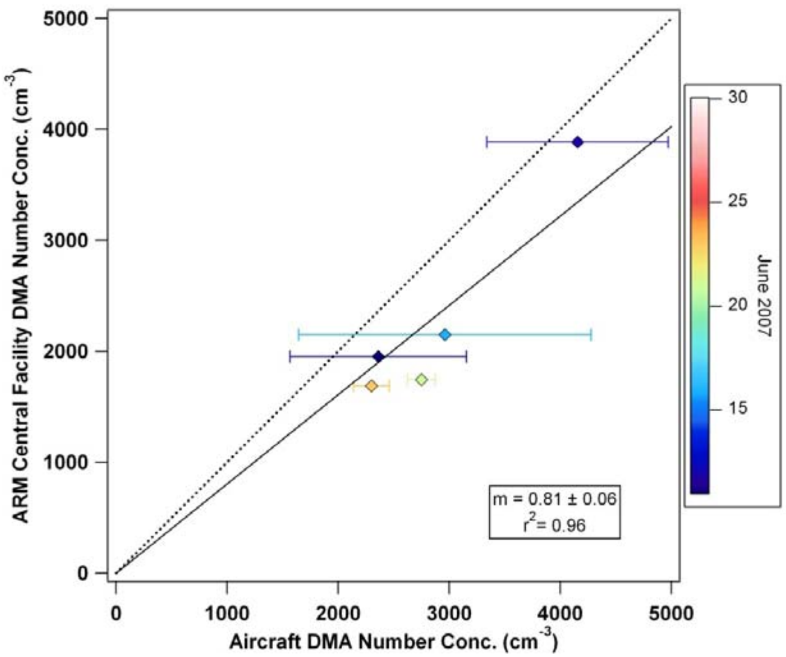
$$\frac{d\left(e'/e_s\right)}{dD_p} = 0$$



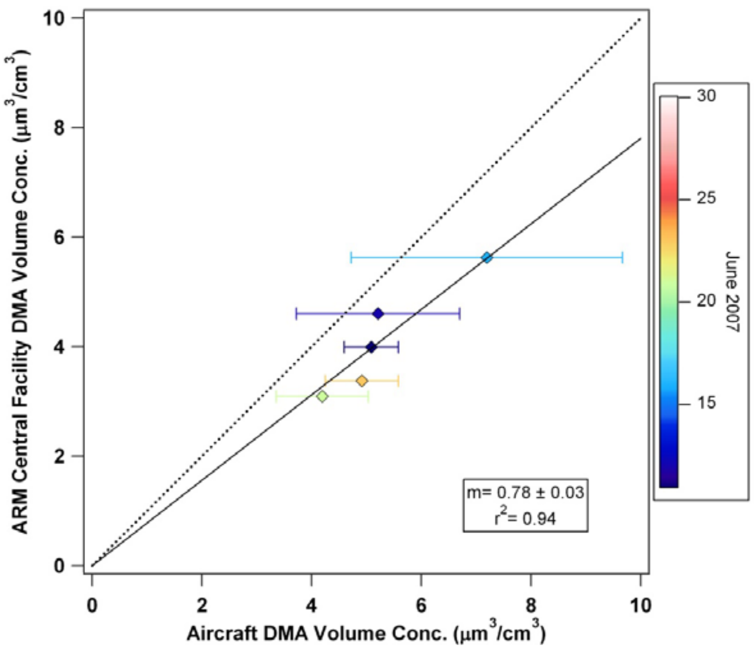
TDMA Köhler Equation → Mass of Solute → Köhler equation #2 → Köhler Curve → Obtain S_c



- Comparison between integrated number concentration and measured concentration from the AOS TSI 3010

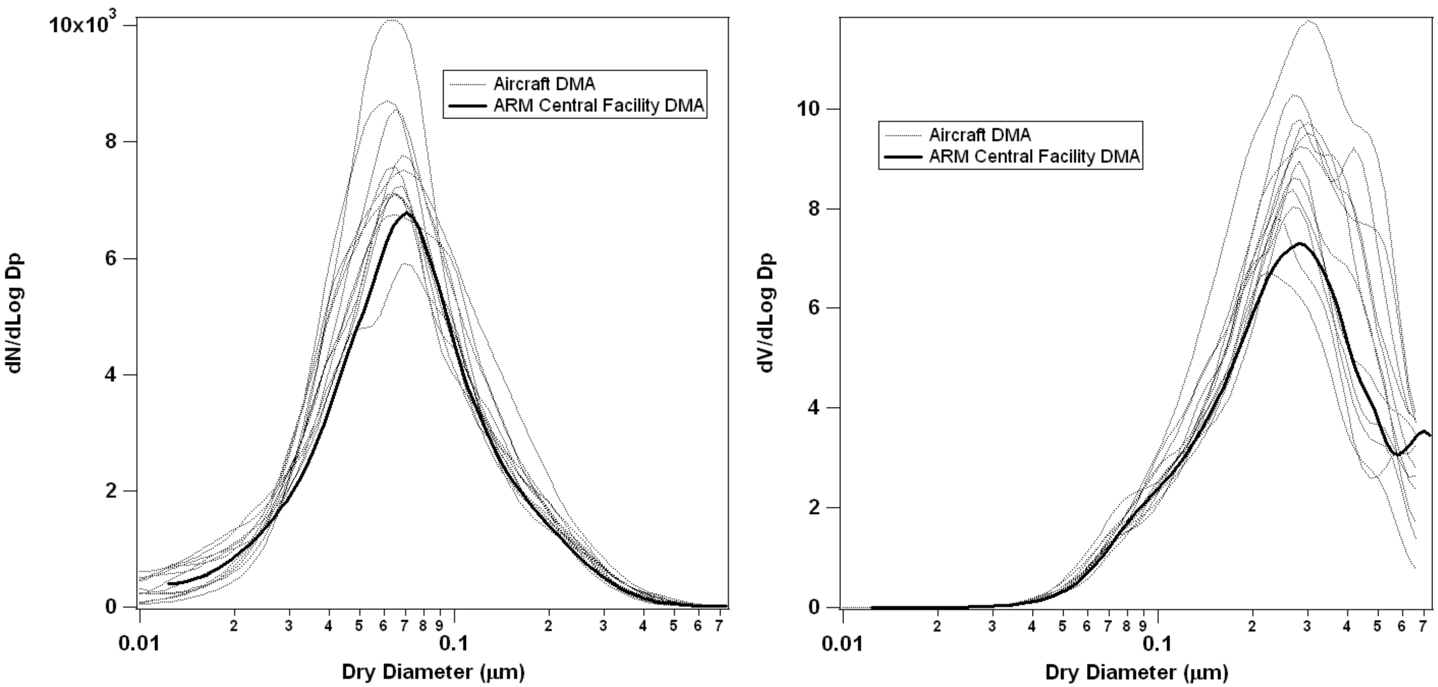


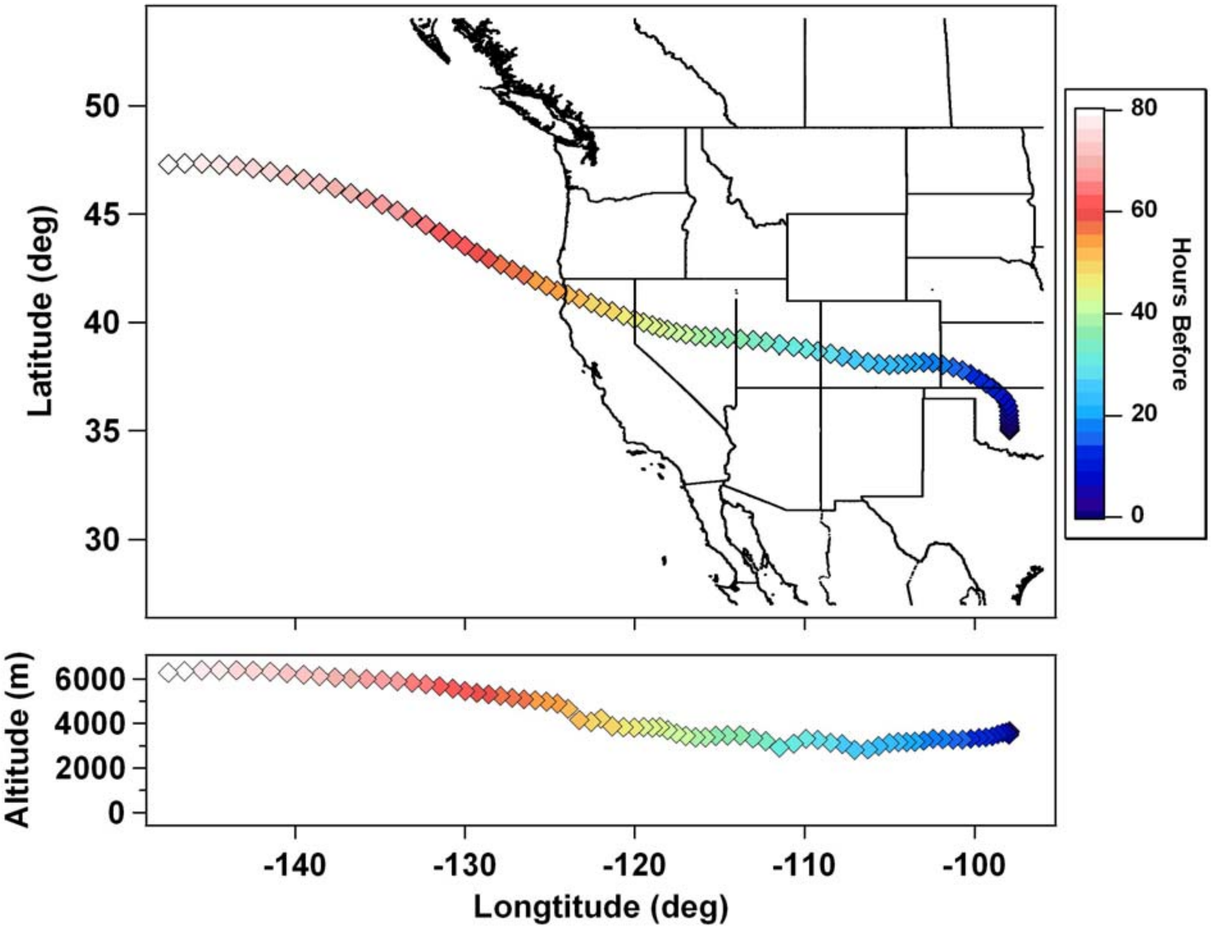
5000
4000
3000
2000
1000
0
ARM Central Facility DMA Number Conc. (cm-3)
0
1000
2000
3000
4000
5000
Aircraft DMA Number Conc. (cm-3)
m = 0.81 ± 0.06
r2= 0.96
30
25
20
15
June 2007



10
8
6
4
2
0
ARM Central Facility DMA Volume Conc. (µm3/cm3)
0
2
4
6
8
10
Aircraft DMA Volume Conc. (µm3/cm3)
m= 0.78 ± 0.03
r2= 0.94
30
25
20
15
June 2007

June 12th Morning Flight





Latitude (deg)
50
45
40
35
30
Hours Before
80
60
40
20
0
Altitude (m)
6000
4000
2000
0
-140
-130
-120
-110
-100
Longtitude (deg)