Calibration Uncertainties in Cloud Condensation Nuclei Counters

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Motivation

- Cloud Condensation Nuclei (CCN) play a major role in cloud and precipitation formation
- CCN are an important subset of aersols that nucleate water vapor in supersaturations characteristic of the atmosphere (< 1.0 %)
- CCN measurements are used in weather modification and modeling research
- Valid CCN measurements with uncertainties help in comparing measurements between projects
 - Knowing uncertainties helps as CCN measurements are input into models

Objectives

- Understand the calibration uncertainties and effects on CCN measurements
- Quantify the pressure dependence is helpful in aircraft operations
- Apply these results to past weather modification experiments (POLCAST)



Droplet Measurement Technologies CCN Counter

- Widely used, commercially available
- Frequently used for weather modification research
 - Understanding it's uncertainties will aid in the comparability of measurements
- The DMT CCN counter is a dynamic vertical thermal gradient diffusion chamber
 - A temperature gradient is applied while flow is continuous through the droplet growth chamber
- The inner walls of the chamber are kept wet using an alumina bisque liner and temperatures are controlled at the top, middle, and bottom of the chamber
- Water vapor diffuses more quickly in air than heat and as both diffuse toward the center of the chamber, there is more water vapor than in thermodynamic equilibrium creating a supersaturation



Droplet Measurement Technologies CCN Counter

- The supersaturation at the centerline of the chamber depends on the temperature difference between the top and bottom of the chamber, pressure in the chamber, and flow rate
 - Supersaturation can be varied between 0.1 2.0 %
- The sheath to sample flow ratio determines how confined the sample stream is to the centerline
- The sample flow determines the volume where counted particles reside
- An optical particle counter at the bottom of the chamber detects and sizes the grown droplets



DMT CCN Counter Calibrations

- Three main calibrations
 - Pressure, flow rate, supersaturation
- Pressure calibration
 - Straightforward comparison of voltage to standard measured pressure
 - Important because pressure influences the supersaturation inside the instrument
- Flow rate Calibration
 - Voltage is related to measured flow rate using a standard flow meter
 - Determines accuracy of concentration measurement and constant supersaturation
- Supersaturation calibration requires a complex lab setup and complex processing



- Pressure Calibration changed by less than 1%
- Don't expect much change in pressure calibrations over time
 - Negligible uncertainty introduced through pressure calibration

Sheath Flow Calibration



- Uncertainty measurements taken 6 months after calibration
- With a setpoint of 455 ccm, the ten sample average of measured flow is 443.43 ccm
- 2.5 % error in sheath flow

Sample Flow Calibration



- Uncertainty measurements taken 6 months after calibration
- With a setpoint of 45 ccm, 10 sample average measured flow is 46.94 ccm
- 4.3 % error in sample flow

Lab Setup: Cloud Condensation Nuclei Counter Calibrations



- Generate aerosols of known size and chemical composition
- Introduce into the DMT CCN counter while holding its chamber temperature gradient constant
- CCN are counted while a calibration standard counts all particles simultaneously
- The ratio of CCN concentration to total particle counts is the activated ratio
- Selected sizes are introduced into the CCN counter at regular intervals yielding activated ratios between 0 and 100 percent



Activation size of 70.08 nm results in a calculated supersaturation of 0.256%

- The activation curve is made using a sigmoidal curve fitting routine to fit the data
- The same processing script determines the activation size based on the size at which the activation curve crosses the activation ratio
 - Normalization of ratio data to 1.0 does not significantly impact activation size calculation (< 0.5 %)
- Using kappa-Kohler theory, the critical supersaturation is calclulated
- Critical supersaturation is calculated at 5 different instrument temperature gradients
- Process is repeated three times at each of three pressures: 700, 840, and 980 mb



- Calculated supersaturation is plotted with its corresponding temperature gradient and fitted linearly
- The fit equation coefficients are used as the instrument's calibration coefficients

Supersaturation Uncertainties

• The relative deviation of supersaturation can be calculated at each temperature gradient to give the uncertainty in calculating supersaturation

- 0.1-0.3 % relative uncertainty

- The overall supersaturation calibration uncertainty is calculated from the relative error of the three calibrations at a given pressure
 - 2.3, 3.1, and 4.4 % uncertainty for 980, 840, and 700 mb calibrations respectively



- Observed average pressure dependence of 0.047 % supersaturation per 100 mb
 - Rose et al. (2008) found a pressure dependence of 0.037 % supersaturation per 100 mb at a temperature gradient of 5 K while this research found 0.039% per 100 mb at a temperature gradient of 6 K
- Slope increases 5.4 % per 100 mb decrease in pressure meaning pressure dependence is not constant
- Single supersaturation offset leads to a corresponding error in supersaturation percent between 1-5%

Uncertainty Effects on Concentration Measurements

Supersaturation Spectrum of Outdoor Air



- 2.3 percent uncertainty from the supersaturation calibration and 4.3 percent uncertainty in sample flow
- Based on the supersaturation spectrum with the ambient calibration and the uncertainties found, concentrations will be within 8.8 percent of the measured value
- Assuming the same supersaturation to concentration relationship, measured values will ¹⁵ be within 10.4 percent at 840 mb and within 13.0 percent at 700 mb

Comparison of Calibration Methodologies

- DMT performed a calibration on SN 062 June 2015
- UND calibrations were done approximately one year later
- Performance checks confirmed that all major leaks developed during shipping were fixed before UND calibrations begun

Supersaturation Calibrations at 840 mb



 UND calibrations are 42-45 % lower than DMT calibrations

Possible Differences in Calibration Methodology

- The assumptions made in Kohler theory calculations can dramatically alter calculated supersaturation
- Multiply charged particles can get through the Electrostatic Classifier and influence the activation curves
- Fitting methods other than a sigmoidal fit lead to large error in calculating activation size



 The calculated supersaturation using the DMT variation of Kohler theory is on average 3.3 % lower than when using κ-Kohler theory

June 2015 DMT Supersaturation Calibration

Temperature Gradient of 3 degrees C at 840 mb



- Plateau around 30 % indicates that 30 % of particles are multiply charged
 - Research indicates plateau heights greater than 10% have a significant influence on calculated supersaturation
- Concentrations of multiply charged particles are not constant over the size range making corrections difficult
 - Knowledge of the size distribution is necessary to correct the data
- A sigmoidal fit is necessary to determine the activation size, not linear interpolation
 - Linear interpolation disregards the ends of the activation curve



Temperature Gradient of 3 degrees C at 840 mb



- Plateau heights must remain below 10% if no corrections are made
- Kohler theory variations can significantly differ so it is important to quantify this when doing comparisons
- A sigmoidal fit, rather than a linear interpolation is necessary for accurately and objectively finding the activation size

Conclusions

- Calibrations should be done under the environmental conditions the CCN counter will be operating under for the most accurate measurements due to a changing calibration slope with pressure
- The uncertainty in the DMT's concentration measurement using this calibration methodology is 8.8, 10.4, and 13.0 % for 980, 840, and 700 mb respectively
- Comparisons between the calibrations done at UND and DMT show 42-45 % lower supersaturations for the same temperature gradient using the UND methodology

Future Work

- Publish results
- Apply uncertainties to POLCAST data
- Explore the supersaturation dependence on temperature and flow rate

References

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Importance of Neutralization

Size Selection of 15 nm Without Neutralizer





- SMPS scan of DMA selecting 15 nm particles without a neutralizer installed.
- Each peak corresponds to a distinct charge state. 15 nm is +1, 21 nm is +2, 26 nm is +3, and so on.
- 5 distinct peaks representing 5 charge states. This indicates the charge distribution of generated particles is much wider than previously thought.
- 51.6 % of all particles in this scan are not within 2.5 nm of 15 nm.
- Electrostatic Classifier manual notes that aerosols can accumulate inside the neutralizer. If it is not older 25 than Kr-85 half-life of 10.7 years, then accumulation is a possible reason the neutralizer may lose effectiveness (thin layer can block alpha radiation).

Kappa-Kohler Theory

- The equation developed by Petters and Kreidenweis (2007) is: $S(D) = \frac{D^3 - D_d^3}{D^3 - D_d^3(1 - \kappa)} \exp\left(\frac{4\sigma_{s/a}M_w}{RT\rho_w D}\right)$
 - Where kappa can be defined as:

$$\kappa = \frac{4 A^3}{27 D_d^3 \ln^2(S_c)}$$

- And A is defined as:

$$A = \frac{4 \,\sigma_{s/a} M_w}{RT \,\rho_w}$$

- Again, ρ_w is the density of water, M_w is the molecular weight of water, σ_{s/a} is the surface tension of the solution/air interface, R is the universal gas constant, T is temperature, and D is the diameter of the droplet. D_d is dry particle diameter and S_c is the critical supersaturation corresponding to the dry diameter
- Kappa for ammonium sulfate is 0.61