

## **Cold and Transition Season Cloud Condensation Nuclei Measurements in Western Colorado**

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Reviewed by Jeff Snider, University of Wyoming

### **Overview**

Ward and Cotton present airborne measurements of cloud condensation nuclei, accumulation mode aerosol and condensation nuclei. Measurements were made upwind of the continental divide in northern Colorado where observations and modeling indicate that inhibition of snowfall occurs with increasing aerosol concentration. Measurements from this region are needed for improved understanding of the aerosol properties and for model validation.

I feel that the manuscript is too “rough” to be put through to ACP with minor revision. As I elaborate below, there are many technical errors. Given the stature of the scientists involved, I am certain that these can be patched up, but troubling for me is the tone of key parts of the presentation. Yes it is true that instrument bias is lurking in the data - it always is. But before going to that conclusion the fine points need to be sharpened, and most important, the analyst needs to examine and double-check assumptions about the data and how it is compared.

Most evident in this regard is the closure study. This starts on difficult footing because the smallest SS, assuming a reasonable value for kappa, corresponds to a particle size which is substantially smaller than the minimum size detected by the optical particle counter. While I see merit in doing the closure study with kappa = 0.1, it needs to be recognized, from the get-go, that the selected kappa is likely a factor 3 to 5 smaller than indicated by the aerosol chemistry. In my opinion, the departure of assumption (kappa=0.1) from reality (PM2.5 chemistry) reflects that CCN prediction is rather insensitive to chemistry and more sensitive to measurements of the cumulative aerosol concentration (either from an aerosol spectrometer or a CCN instrument) and how those data, and their uncertainties, are accounted for in the closure comparison.

### **A Proper Error Analysis is Needed for the Closure Study**

In Snider et al. (2006) we report on uncertainty in the measurement of CCN concentration, and uncertainty in the chamber supersaturation. These are quantified and discussed. Further, we demonstrated that the former has contribution “...from the random placement of droplets within the scattering volume and from the angular asymmetry of the scattering.” Given that, I feel it is inappropriate to “...sum the Poisson error and 30% additional instrument uncertainty...” More importantly, it was not established that the 30% (set by Ward and Cotton) is the appropriate relative uncertainty for the CCN concentration measurements presented here. In my opinion a proper error analysis, based on the information presented in Snider et al. (2006) is needed. The SS uncertainty, which affects the predicted  $N_{ccn}$ , also needs to be accounted for in the closure study.

## General Comments

There are many representations of the supersaturation in this paper (SS, SSnom, SSeff, SSc). The chamber was calibrated correctly, as far as we know, so it would be fair (I feel) to equate SS with SSeff and leave it that way throughout. In my opinion the symbol SSnom should only appear in the discussion of the calibrated relationship between SS (SSeff) and SSnom.

Maybe I missed something, but why is there focus of SSeff = 0.27% when the presentation of the data (Figure 6) shows SSeff = 0.20 % as the minimum measurement supersaturation?

It seems, from Figure 14 of Politovich and Vali (1983), that sensitivity of droplet number concentration to the Twomey  $k$  is significant, particularly at relatively large  $C$ . As far as I can tell this paper does not provide recognition of that, nor does Saleeby et al. (2009). Saleeby et al. (2009) change  $N_{ccn}$ , but it would seem that change in  $k$  (slope of the cumulative aerosol or CCN spectrum), particularly at large but not unreasonable values of  $C$ , might have a more significant impact on droplet concentration than  $C$  itself. Also, Saleeby et al. (2009) assume the particles are ammonium sulfate. Needless to say there is a significant difference between the RAMS-assumed aerosol chemistry and the “closure” chemistry. The implications of these two observations, *visa a vis* the RAMS model, should be discussed.

On page 27656 is “ambient SS” the correct supersaturation in the particular context? This comes up in a few other places: “Cloud-active particles” and “CCN-active aerosol” should be defined.

## Specific Comments

### P27634 L10

Politovich and Vali (1983) is overlooked as a reference for prior CCN measurements; perhaps because the upper North Platte valley is not a “Western Slope” location. In my opinion there are many similarities of the two locations, most importantly, both are “rural” yet both have significant upwind point sources within 100 km (Sinclair Refinery and further west Bridger). Also relevant, but perhaps not cited for the same reason, is the lower tropospheric aerosol climatology (CCN, CN, larger particles) from over Laramie (Delene and Deshler, 2001).

### P27642 L18

The authors assert that no supersaturation (SS) is specified in PV83. Yet the parameters describing the activation spectrum are plotted (their Figure 14), and from those values the  $N$  at any SS is derivable.

### P27638 L4

Is this sentence necessary?

### P27638 L16

Only one measurement every 10 s for the CPC? This is different on the King Air, where we sample the analog output signal at 1 Hz. Should this distinction be pointed out?

### Table 1

The ratio  $C3/C5$  should be less than unity. But it seems that this should be the effective supersaturation (SS=SSeff) which from Figure 6 is the ratio of the concentration at 0.20% to 0.43%.

**P27644**

As you point out, the CCNC-100A does not actually “count” rather it measures the intensity of the light scattered by an ensemble of particles. For that reason, I feel it is confusing to refer to the CCNC-100A as a “CCN counter”. I prefer “CCN instrument.” Back in the day, the illuminated droplets were photographed, so that was the reason for the final “C” in CCNC. Also, within the chamber droplet growth occurs via the process of vapor diffusion – the droplets hardly “diffuse.” Finally, I feel that there is an important distinction to be made between SS, an experimental variable, and SSc, a property of a particle. Hence, I recommend that “Particles that require a SS below...” be changed to “Particles with a critical supersaturation (SSc) less than...”

**P27644 L13**

The chamber is “circular” in one dimension, but the way it is stated here I had an image of a “spherical” chamber.

**P27645 L1-6**

The flush interval and detection intervals are fixed (total is 25 s), but the time for temperature adjustment can be as long as 15s, particularly if there is a large change in the plate temperature between measurements. This paragraph came across as if there was something not working the way it is supposed to.

**P27645 L15**

Please check what the actual SSnom values were for your measurements (on average). It is likely that your averages are significantly different from the values you state here. This is important because SS=0.27% is used in the closure, but Figure 6 indicates that this should be more like SS=0.20%.

**P27645 L15-16**

Some caution is needed here. Prior to 10 December 2009 the coefficient that converts from nominal to effective supersaturation was different. After installing a new thermocouple (December 10 to 11), and finishing the project, we did a second CCN calibration for ISPA09. Here are coefficients from the NetCDF files:

```
[jsnider@bat processed]$ ncdump -h 20091210a.c1.nc | grep DTEMP_RAW:SSCal
DTEMP_RAW:SSCal = 0.61 ;
DTEMP_RAW:SSCalibrationDate = 2009. ;
```

```
[jsnider@bat processed]$ ncdump -h 20091211a.c1.nc | grep DTEMP_RAW:SSCal
DTEMP_RAW:SSCal = 0.7 ;
DTEMP_RAW:SSCalibrationDate = 2010. ;
```

### **P27645 L26**

By my reckoning there are 28 channels with diameter larger than 0.134 nm, i.e., 0.134 um is the upper limit size of the second of thirty channels. Note below that the sizing array is 31 values for 30 channels of OPC data, and that of the 31 bins of data the “Bin 0 is empty (compatibility). Bins 1-30 contain accumulations from probe.” Because of this possible misinterpretation, the concentration/size relationship you use in the closure may need to be revised. Here are the details from the NetCDF file:

```
[jsnider@bat processed]$ ncdump -h 20091211a.c1.nc | grep AS200
float AS200_OBL(time, sps1, Vec31) ;
  AS200_OBL:long_name = "PMS PCASP number (per cell)" ;
  AS200_OBL:units = "number" ;
  AS200_OBL:Category = "PMS Probe" ;
  AS200_OBL:SampledRate = 1 ;
  AS200_OBL:FirstBin = 2 ;
  AS200_OBL>LastBin = 30 ;
  AS200_OBL:VectorLength = 31 ;
  AS200_OBL:Package = "get_pcasp1" ;
  AS200_OBL:matlab_name = "NPcasp" ;
  AS200_OBL:CellSizes = 0.108f, 0.122f, 0.134f, 0.143f, 0.152f, 0.16f, 0.168f, 0.176f, 0.185f, 0.197f,
0.21f, 0.227f, 0.247f, 0.273f, 0.303f, 0.339f, 0.426f, 0.57f, 0.721f, 0.88f, 1.047f, 1.222f, 1.405f, 1.596f, 1.794f,
2.001f, 2.215f, 2.437f, 2.667f, 2.905f, 3.151f ;
  AS200_OBL:OutputRate = 1 ;
  AS200_OBL:SerialNumber = "1013-0502-29" ;
  AS200_OBL:comment0 = "Bins are indexed 0 - 30 " ;
  AS200_OBL:comment1 = "Measured: 30 bins (1-30);" ;
  AS200_OBL:comment2 = "Bin 0 is empty (compatibility). Bins 1-30 contain accumulations from probe;" ;
  AS200_OBL:comment3 = "Bin 1 is noisy, thus usually omitted;" ;
  AS200_OBL:comment4 = "FirstBin=2, LastBin=30; Those bins are used for concentration calculations;" ;
  AS200_OBL:comment5 = "CellSizes are bin edges for bin 1-30 (lower edge of bin 1 is uncertain)."
```

### **P27646 L6-7**

Since the detection is different for the CCN (measures scattering from an ensemble), versus the PCASP (single particle detection), this sentence is confusing. I contend that both instruments detect all particles they sample, but you appear have another definition for the word “sample.” Please clarify.

### **P27646 L8-9**

Crossing the detector beam is not a necessary condition for detection. Rather, an ensemble of droplets, within the scattering volume, produce scattering which is detected and related to concentration via the detector calibration (Snider et al., 2006). Late in the 20 s interval, and if the  $SSc < SS$  condition is met, a subset activate and begin to fall. Sedimentation is the reason the scattering signal diminishes from a peak value (a lower concentration falls from above the laser beam). This is explained in both Delene and Deshler (2000) and in Snider et al. (2006). I am critical of what you wrote here. In my opinion you are mixing the issue of Poisson counting, for the CCN and PCASP, with explanation of how the Wyoming instrument detects CCN. I advise that these topics be separated. With regard to the relevance of Poisson counting to the Wyoming CCN, we probed the reason for the variability in the detector calibration, and showed that the observed concentration variability is consistent with the prediction of Poisson statistical theory (Snider et al., 2006). From scattering theory, there was also indication that where the droplets position within the viewing volume also influences the variability. There is also discussion of this in Snider et al. (2006).

**Table 2**

Flight duration is in excess of five hours. Were there two flights, of approximately 3 hour duration, on each of these days?

**P27647 L27 into P27468**

I don't see where you explain that the scattering peak voltage ( $V_{\text{peak}}$ ) is the value used to deduce the CCN concentration, via the detector calibration. Once that is explained, the data QA discussion makes more sense to the outside reader. Did Oolman or Wechsler inform about the details of the QA checks. In this context, what do you mean by "noise profile", "minimum" and "maximum"? Perhaps it would be good to report the fraction of measurements at each SS that were valid, on a project-wide basis.

**Figure 6**

The caption states that error bars (one standard deviation) are plotted. The error bars are not evident. I recommend that the individual data points not be shown, rather, I suggest that you show the averages and their standard deviations. Also, a log-vs-log plot would be useful for those interested in C/k parameterization of activation spectrum (power function of the form  $N=C*SS^k$ ). Finally, I recommend that in Figure 6 you show the PCASP average concentration ( $s$ ) and standard deviation. That would provide a good introduction for what follows in Figure 9, i.e., that there is not "perfect" agreement (assuming a kappa which is smaller than indicated by the aerosol chemistry) with the assumed chemistry but on average there is fair agreement.

**P27648 L24-25**

The two-times larger active fractions at MVNP, which you refer to, were measured with the DMT CCN instrument at ground level? I advise that you be more specific about this, because as it is stated here it is not clear if MVNP is indicating airborne or surface data. If it is MVNP surface data that is being referred to that could imply negative bias in the CCN measurement made with the Wyoming instrument or positive bias in DMT. Have you thought about this? Are the DMT values expressed at ambient temperature and pressure, or extrapolated to sea level?

**P27648 L29**

I am not sure what is the meaning of "...can be disrupted by sharp changes in ambient air pressure..." and "...particle detection is delayed."

**Figure 8**

What is said in the caption ( $N(SS_{\text{nom}}=0.8\%)$ ), and in the text ( $N$  at all three  $SS_{\text{nom}}$ ), are inconsistent.

**P27652 L7-L10**

It is not clear why temperature is used. If you are using the temperature as input into the Kelvin term of the PK07 Koehler equation, I would argue against that. If you do, then I would want to know how you parameterized the temperature-dependent surface tension and the temperature-dependent water in the Kelvin portion of the Koehler equation. My opinion is that there is insufficient information about the temperature dependence to warrant consideration of these important, but poorly characterized effects. For more discussion of this, consult Section 4.5 of this paper: Snider et al., JGR, 115, D11205, 2010. My recommendation is to fix the Kelvin parameters ( $T$ , surface tension and water substance density), at standard values recommended by PK07. Related to this, I don't see the relevance of the equations that were presented in the explanation of PK07. What is needed, for your analysis, is a statement of the mathematical expression for the Koehler curve (from PK07) and a discussion of what the parameters are and what was assumed for their temperature dependence. Given that the word "hygroscopicity" is commonly used to communicate growth as a function of RH, I don't think it is relevant relevance in this section.

**P27653 L7**

The minimum detectable size plus the assumption that the particles have an index of refraction equal 1.59. This refractive index assumption needs to be put up in the experimental section. As I mentioned earlier, this size is incorrect.

**P27653 L24**

What was done with points that had measurements of N at the smallest SS greater than the size-integrated PCASP concentration? Were there many of these?

**P27653 L9**

critical \_dry\_ diameter of 0.07 um

**P27654 L4-L5**

I don't agree with this conclusion. First, you were "forced" to use  $\kappa = 0.1$  because larger values would make it impossible to do the closure comparison (critical dry size smaller than the minimum size detected by the PCASP). Second, the  $\kappa$  you picked is inconsistent with the aerosol chemical composition data, showing that at least half of the mass corresponds to  $\kappa \geq 0.6$  materials. You make a good rebuttal to this criticism on page 27655 – the larger particles determine the chemistry. But why should their composition be so different (not much soil dust in the wintertime) from the composition (at smaller sizes) that is relevant to the aerosol-to-CCN closure?

**P27654 L2**

I can see no justification for summing the Poisson error for each size bin. Better would be the standard deviation of the 20 PCASP concentrations corresponding to the concentration of "...particles in size bins greater than the critical (dry) size.."?

**P27652 L4**

"Since composition measurements were not taken during this project,..." The PM2.5 measurements do not qualify here?