

## ***Interactive comment on “Cold and transition season cloud condensation nuclei measurements in western Colorado” by D. S. Ward and W. R. Cotton***

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Thank you, Dr. Snider, for your comments and critique. We have addressed all the recommendations point-by-point here. These comments not only greatly improved the manuscript but also improved our understanding of the KingAir instrumentation.

General Comments 1. 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.

C13959

This is a good suggestion; it improves the clarity of the paper and would prevent some potential confusion. A sentence noting that the SSeff will be referred to as the SS for the KingAir section was added to the text.

2. 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?

This was corrected in the text. In accordance with the above comment, the SS (SSeff) averaged about 0.22% for the project and this value was used as the focus in the revised text.

3. 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.

We felt that issues of size distribution representation in modeling of cloud microphysics, while important (likely more important than composition representation) were for the most part outside the scope of this paper. Some droplet activation parameterizations allow for different size distribution shapes, but many others (including RAMS) use a lognormal representation of the aerosol size distribution. The inaccuracy potentially introduced by this assumption is noted by Ward et al. (2010 – ACP). We do include mention of the size distribution (expressed in this case as the portion of total particle number comprised by Aitken-size non-activating particles) as the likely cause of the

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variability in Ncn that is not matched by the variability in Nccn over time.

In addition, while Saleeby et al. (2009)'s use of ammonium sulfate differs from the closure chemistry assumed here and from the general conclusions of regional aerosol observations, they were testing the sensitivity of orographic clouds and precipitation to CCN number concentration only. Certainly changes in the assumed aerosol composition will change Nccn, but for the sensitivity studies one uses a constant composition and there is flexibility in the choice of the composition as long as it leads to realistic Nccn, or the desired Nccn within the model. Because of this, Saleeby et al. (2009) might not be a good benchmark for comparing against the chemistry assumed in the current study. The published observations of aerosol composition from nearby sites provide a better comparison; these are highlighted in the revised manuscript in accordance with the suggestions in this review.

4. 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.

Cloud-active particles were defined at P27634, L15:

"...the fraction of the aerosol population that will nucleate a cloud droplet for a given environment..."

And references to CCN-active were removed for simplicity.

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).

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Reference to these studies was added at this point in the text, the tone of the original text did make it seem like a larger region was void of this type of measurement.

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.

We estimated the SS for 7 points from Figure 14 of PV83. All points were taken from on or very near to a line of constant N since the scale of N in between the lines is not shown but is probably not linear (judging by the slopes of the other N lines). Using the Twomey (1959) power law equation we estimate SS between 0.4% and 0.65% for the observations made by PV83. This is indicated in the text.

P27638 L4 Is this sentence necessary?

The sentence was superfluous – it was deleted.

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?

The CPC did sample every second, we only recorded the data every 10 seconds at MVNP and mis-represented this in the text. We changed the text to show that the CPC at MVNP was also sampling every second but we only used one out of every ten obs.

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%.

This was a typing error, the ratio was changed to C5/C3 to match the data in the table. Since these data are from the DMT CCN-100 the SS are reported as the value given in the instrument setting.

P27644 As you point out, the CCNC-100A does not actually "count" rather it measures

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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. . ."

All these recommended changes were made.

P27644 L13 The chamber is "circular" in one dimension, but the way it is stated here I had an image of a "spherical" chamber.

We noted that the chamber is circular in the horizontal dimension.

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.

The text was changed to communicate this better: P27645 L2-6 "The entire process takes 30 to 40 s including the time needed to flush the old sample out of the chamber, admit the new sample, and achieve the steady state SS which can be as much as 15 s depending on how long it takes for the temperature gradient within the chamber to stabilize (Delene and Deshler, 2000; Snider et al., 2006)."

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%.

The SSnom and corresponding SSeff values were averaged for the campaign and the

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average SSeff were reported in the text – 0.22%, 0.46%, 0.96%.

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. ; [js-
nider@bat processed]$ ncdump -h 20091211a.c1.nc | grep DTEMP_RAW:SSCal
DTEMP_RAW:SSCal = 0.7 ; DTEMP_RAW:SSCalibrationDate = 2010. ;
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This is mentioned in the added discussion of the chamber calibration in section 3.1.1 (see response to comment P27646 L8-9).

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

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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).";

This was a typo only, albeit a significant one. The number 2-30 bins were used in the analysis. The typo was fixed in the text.

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.

See next comment:

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

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the variability. There is also discussion of this in Snider et al. (2006).

The discussion of the Poisson error that was commented on here and in the previous comment was removed from the text. The topic of the detector calibration and uncertainty were included in several new paragraphs starting in Section 3.1.1 (renamed 'Instruments and calibration) that follows the work of Snider et al. (2006) and Snider et al. (2010). The following major portions of text replace the existing text:

"A laser illuminates the center of the chamber (assumed to be the region of maximum SS) where the light is scattered by the particles present in the chamber. The amount of scattering is monitored by a photodetector and can used to derive the number concentration of large particles (droplets) in the chamber. During the detection stage the chamber SS may reach a maximum as activated droplets grow in the supersaturated environment and deplete the available water vapor. The photodetector voltage often reaches its peak value at this point, when the droplets are at or near their maximum number and size. The peak voltage is used to determine the Nccn. The relationship between the light scattering, as indicated by the peak photodetector voltage, and the droplet number concentration is calibrated by testing an aerosol population with a 100% activation rate and known number concentration (Snider et al., 2006). The variability in the resulting peak voltage measured for test aerosol populations with the same Nccn is used to estimate the uncertainty associated with the CCN instrument detector as shown by Snider et al. (2006)." And:

"The  $SS_{eff}:SS_{nom}$  ratio and associated uncertainty are estimated during chamber calibration by testing the activation behavior of size-selected ammonium sulfate particles (Snider et al., 2010). The true chamber SS can be determined from the activation behavior of the ammonium sulfate and a Köhler theory model (Snider et al., 2006). This is compared to the theoretical  $SS_{nom}$  to give  $SS_{eff}:SS_{nom}$ . For ISPA-III, the original chamber calibration found  $SS_{eff}:SS_{nom} = 0.61$ . During the project a plate temperature sensor failed and the result of a new calibration ( $SS_{eff}:SS_{nom} = 0.7$ ) was retroactively applied to data collected after the failure (Dec. 11 through Dec. 19). Using results from

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Snider et al. (2006) the chamber calibration uncertainty is estimated to be 10% of SS-eff:SSnom for the current study. It is assumed in the following analysis that the SSeff represents the true SS experienced by particles within the chamber and will be referred to simply as SS for the remaining discussion of this instrument and its measurements.”

Table 2 Flight duration is in excess of five hours. Were there two flights, of approximately 3 hour duration, on each of these days?

The following text was added to clarify this point:

P27647 L20 "Each flight day contained two flights with a stop at a southern Colorado airport for refueling. These airports were Cortez (CTZ) for flight plan #1 and Montrose for flight plan #2 (MJT; see Figure 1)."

Table 2: Note the flight duration is the total time from the southward and northward flights from each day.

P27647 L27 into P27468 I don't see where you explain that the scattering peak voltage ( $V_{peak}$ ) 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 Colman 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.

Mention of  $V_{peak}$  is now included (see response to P27646 L8-9), but since the very few details that were given on the  $V_{peak}$  quality checks in the manuscript could have been confusing, these were left out. We did add mention of the fraction of measurements at each SS that passed the quality checks. These were 0.55 (SS=0.22%), 0.76 (SS=0.46%) and 0.85 (SS=0.96%).

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,

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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.

Figure 6 was updated to include the standard deviation error bars and the PCASP information. Note that the PCASP concentrations for bins 2-30 were included. The CCN means and sd's were plotted on a log-log scale. The figure was difficult to understand when plotted with all the flow regimes on a single plot so each regime was given its own window and the mean of all data regardless of flow regime was emphasized in a larger window. The text was altered to reflect these changes and the caption was re-written.

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?

The DMT values are output at ambient pressure and temperature (if the instrument temperature is considered ambient). We added reference to this on pg. 27637, L21. The comparison between the airborne KingAir CCN observations and the ground-based MVNP observations is difficult and, as you note, the differences could result from several factors which aren't elucidated here. Therefore, this sentence was removed.

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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."

These sentences were removed.

Figure 8 What is said in the caption (N(SS<sub>nom</sub>=0.8%), and in the text (N at all three SS<sub>nom</sub>), are inconsistent.

This was fixed in the text and in the caption. The figure shows vertical profiles of N<sub>ccn</sub> measured at SS = 0.46% for both the southern and northern stack locations.

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.

The computation of the critical radii for the CCN closure study was redone with the PK07 temperature, surface tension and water density. Reference was made to the discussion in Snider et al. (2010) on this topic. Also, Eq.'s 2,7 from PK07 were removed and Eq. 6 was included instead. You're right; this is much more relevant as it is the equation used to compute the critical radius. We removed references to the hygro-

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scopicity of particles in the section and used SSc or kappa to characterize them in this context.

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.

The assumed refractive index was added to section 3.1.1. The size error was fixed both here and in section 3.1.1. Bin 0 is empty and bin 1 has an uncertain lower size limit so we used the concentration data from bin 2 and up for the closure study.

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?

These data points were set to missing. There were a total of 41 such data points which is less than 2% of the total valid CCN data points.

P27653 L9 critical dry diameter of 0.07 um

Changed in the text.

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 ≈ 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?

This conclusion was played down as one possibility in three, and perhaps the least likely of them. The new text:

"There are several possible explanations for this unexpected result. First, the ambient

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aerosol during this project could have been dominated by organic compounds or dust, which are characterized by low hygroscopicity. This conclusion is at odds with the IMPROVE dataset and other published observations in the region. If the ISPA-III ambient aerosol hygroscopicity was more consistent with these previous composition observations, it is possible that the Nccn measurements were simply insensitive to variations in aerosol composition. However, parcel model results suggest that at the low SS used here some sensitivity could be expected (Ward et al., 2010)..."

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.."?

This was done away with by the new error analysis. Instead the standard deviation was used, as suggested here, and the results of the detector and chamber calibrations were also used to compute the error bars for the predicted and observed Nccn.

P27652 L4 "Since composition measurements were not taken during this project,..."  
The PM2.5 measurements do not qualify here?

The text here was changed to note that no composition measurements were taken "in flight". In the end, the PCASP size range dictated the choice of kappa, but a comparison of the chosen kappa against the IMPROVE data was given in the same place where kappa=0.1 was first written.

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 27631, 2010.

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