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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

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 for your comments. We have addressed them here with changes to the manuscript text and figures.

1. Page 27633-27634 - I think it is important to place the issue of anthropogenic effects on precipitation in perspective. For example, Zhang et al., 2007 (Detection of human influence on twentieth-century precipitation trends, Nature, 448, doi:10.1038) estimate that anthropogenic forcing contributed significantly to observed increases in precipitation in the Northern Hemisphere mid-latitudes. That result contrasts with our aerosol particle theories that suggest that precipitation initiated via collisioncoalescence should

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decrease with increasing pre-cloud aerosol. But this raises another issue. Is the orographic precipitation initiated through the ice phase and, if so, how are the CCN important?

The following text was added to the introduction to address this point:

P27633, L12 "Despite numerous studies on this topic, there is considerable uncertainty regarding the effect of anthropogenic aerosols on global precipitation. Global climate model simulations in general predict that increasing aerosols has suppressed precipitation globally (Levin and Cotton, 2009). However, Zhang et al. (2007) reported that precipitation increased in the Northern Hemisphere mid-latitudes during the latter half of the 20th century, suggesting that the role of anthropogenic aerosols is poorly understood. In fact, the magnitude and even the sign of the feedbacks of aerosols on clouds and precipitation depend on the cloud regime being affected and in many cases are not well known (Stevens and Feingold, 2009), making studies of CCN variability important on local and regional scales."

And:

P27633, L15 "In mixed-phase orographic clouds, where ice crystals co-exist with water droplets, it is hypothesized that a decrease in the size of supercooled cloud droplets reduces the riming efficiency, thereby decreasing the liquid water content of precipitation falling as snow or changing the spatial distribution of the precipitation (Borys et al., 2003; Saleeby et al., 2009)."

2. Page 27634 - Lines 7-9 – Elaborate.

These lines in the manuscript did need improving. The text was changed to better explain the reasons for the difference in susceptibility:

"For example, it has been hypothesized that precipitation from high LWC mixed-phase orographic clouds may be more sensitive to increases in aerosol because of the greater potential for riming compared to low LWC clouds that contain more ice (Borys et al.,

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2000). High LWC clouds are more common in southwest Colorado than in northwest Colorado and, thus, may be more susceptible to decreases in riming efficiency due to increases in CCN number concentration."

3. Page 27635, lines 9-15 – Why not indicate the power plant positions in Figure 2. Also, figure 1 needs a map; perhaps an adjacent map of Colorado indicating where this figure is positioned.

We think the reference to Figures 1 and 2 were meant to be switched in this comment. Assuming this is the case, we added the positions of the major power plants local and "upwind" of the MVNP site. This included the three plants in the Farmington, NM area, the Navajo plant in AZ and also the Cholla plant in AZ which we had misnamed in the text (we corrected this).

We also indicate that MVNP and MEVE are co-located on the new Figure 1 to illustrate the location of the IMPROVE site at which the data in Figure 2 were collected.

4. Page 27636, lines 2-4 – The IMPROVE filters are only sampled for 24 hours, once every three days, so why would you show the centre of the 3 days rather than just the day of the sample?

The reason we did it this way was poorly explained in the manuscript so we added some text that hopefully will clear it up. The basic idea is that, yes the filters sample for only one out of three days, but this date is not the same from year to year. Since we are showing ten-year averages it worked best to define three-day periods during which there would be one observation from each year and plot these centered on the middle date of the period.

5. Figure 3 is nice, but it would also be helpful to have the same figure repeated with a linear y-axis to make the relative changes more evident.

We used a log scale originally because the difference between the CN maximum and CCN maximum number concentrations was enough that the variations in CCN could

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not be discerned. But we agree that the relative changes within each time series are hard to interpret on a log y-axis. As a solution we include two images in Figure 3 as recommended by the referee with both on a linear y-axis. The CN and CCN are separated. This better highlights our point that the CN time series exhibits greater variability in time and was an excellent suggestion.

6. Page 27640, line 7 – "used" for?

Added ". . .to represent the large-scale wind in this analysis."

7. Figure 4 – The caption refers to the 500 mb height, whereas in the text it says the 700 mb height (page 27641, lines 10-25)?

Good catch, the correct height was 500 mb so we changed this in the text.

8. Page 27640 – Simply labelling Section 2.3 as "Regression analysis" is not very informative. Rather, something like "Particle-Geopotential Height Regressions"?

We opted for "Aerosol and geopotential height regression analysis".

9. Section 2.4 – The particle regressions with geopotential height are interesting, but no physical interpretations are offered and I am a little disappointed that this analysis is not carried a bit further. Although not significant at the 95% level, higher CCN are associated with higher pressure, whereas the association of CN with pressure is apparently the opposite. The motivation of this paper is the potential impact of the aerosol on precipitation but of course precipitation is a major factor in the reduction of aerosol in the atmosphere as well. Are these patterns related to precipitation? E.g. the removal of CCN by precipitation might lead to the positive correlation(?) and the probability of the formation of smaller particles, measured as CN, after those larger condensational sinks are removed would increase. Are back trajectories consistent with these observed correlations?

We expanded our analysis of the relationship between the particle sampling at MVNP and the large-scale meteorology. In this analysis we consider cloud water, and precipi-

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tation, which is an excellent suggestion. Also we include here the results of a composite analysis on the MVNP local winds that was carried out but not reported originally. The goal is a better understanding of why the CCN and CN might be related to the particular geopotential height pattern as demonstrated in the manuscript.

We started with a back trajectory analysis of air parcels originating at the MVNP location during extreme high and low CN and CCN events that were selected from the CN and CCN timeseries. Four minimum events (9/24 06:00 MST, 9/30 00:00 MST, 10/4 17:00 MST, 10/7 06:00 MST), and three maximum events (9/25 18:00 MST, 10/3 12:00 MST, 10/8 20:00 MST) were chosen. During these events CN and CCN increased or decreased together (activated fraction was relatively constant). Trajectories were computed using the HYSPLIT model with the 40km EDAS meteorological dataset. They were run for 72 hours back from the time of the event. The trajectories were then compared to NARR 3-hr precipitation fields at a time of 24 hours before the CN maximum or minimum at MVNP. In all events it seems likely that the air parcels encountered little to no precipitation. This is shown in figures 1, 2 of this response (included in the response but not in the revised manuscript) which are plots of the NARR precipitation and average HYSPLIT back trajectories for each event. On the whole the study period was very dry also at MVNP with only 3 rain events and a total of 0.65” of precipitation during the entire study period. As the reviewer notes, precipitation is a major removal mechanism for aerosols. In this case, however, it probably did not play an important role in the minimum aerosol events at MVNP.

To get a better idea for wet removal effects on the entire timeseries a regression analysis, similar to that done for geopotential height, was performed for total column cloud water, and 3hr forecast cloud cover in percent (NARR dataset). This analysis did not result in any statistically significant regression relationships or any regions with a consistent sign of the regression coefficient. Since precipitation totals were quite low in the western United States during this time, we conclude that precipitation could be a driver of CN and CCN at MVNP but this cannot be determined from our dataset.

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The back trajectories do show similar air parcel paths for the events in each category – maximum and minimum. The geopotential height regression in the original manuscript shows the relationship between aerosols at MVNP and upper level wind. To include local surface effects, the results of a composite analysis of the CN and CCN with MVNP surface wind direction are shown. By dividing the CCN and activated fraction measurements into wind direction bins it is shown that the greatest CCN number concentrations at MVNP occurred under E-SE surface winds. The activated fraction, however, was the least under these wind directions. This analysis was not included at in the original manuscript because none of the bin means are statistically different from one another at the 90% confidence level (or even much lower confidence levels). But they do support the conclusions drawn from the regression analysis and are interesting even if they do not apply outside the study period.

From this we conclude that the relationship between the geopotential heights at 500mb and MVNP aerosols is largely a result of the flow that is forced by the height pattern and less so because of precipitation associated with the height pattern. It should be noted that while the regression maps for CN and CCN (Figure 4) look opposite, both indicate that high CN and CCN number concentrations are associated with above average heights over the central U.S. and intermountain west. Substantial text was added to the revised manuscript to communicate these points. Also, a figure from the composite analysis was included.

10. Page 27644, line 20-21 – There is more than diffusion going on. Initially, when the chamber is sealed off, there will be some eddies left from the sudden stop of the rapid flow in and out of the chamber. As the eddies subside and the supersaturation profile develops, then the particles will start to grow and there will be some settling. I doubt that particle diffusion is of much consequence in the Wyoming CCNC.

The reference to diffusion was removed from the text.

11. Page 27646, lines 10-15 – Is a Poisson distribution appropriate for droplets growing

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and falling out in a static chamber? Is that a random process?

The random error applies to the detector which, as stated by Snider et al. (2006), will give a range of values for the peak voltage for the same concentration of particles because the droplets are randomly located within a small sampling volume and they scatter more light in the forward direction, meaning the droplets that are far from the detector scatter more light into the detector than similar particles that are closer in. An improved characterization of the instrument error was given in Snider et al. (2006) and so a complete rehashing of the error analysis in this manuscript has been done.

12. Page 27650, lines 12-14 – Does the sampling across 40-60 km truly create a bias? If wind speeds did not change with altitude, then a bias due to the CCN sampling might be a factor, but I assume that your winds varied in speed with altitude and so how do you define a bias in the sampling?

We removed this reference to bias since along-path differences in aerosol quantities should affect all SS settings equally and use of the term "bias" here was misguided. The main idea of this text was that the particle number concentrations as measured by the CPC were not constant across the 40-60 km legs and therefore the CCNC would not always be sampling from the same population of aerosols. This is one explanation for why the CCN number concentrations could be greater at a lower SS at the same altitude.

13. Page 27650, line 24 – An earlier effort at CCN closure was discussed by Liu, P.S.K., Leaitch, W.R., Banic, C.M., Li, S.-M., Ngo, D. and Megaw, W.J., 1996: Aerosol observations at Chebogue Point during the 1993 North Atlantic Regional Experiment: Relationships among cloud condensation nuclei, size distribution and chemistry, J. Geophys. Res, 101, 28971-28990.

Yes this study ought to have been referenced. Added at P27651, L9:

"This was also pointed out in earlier work by Liu et al. (1996) who combined aerosol

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composition measurements with CCN predictions and found that the relationship between particle number and CCN number concentrations depended on the aerosol chemical species being tested."

14. Page 27653, lines 15-20 – This was also shown with a larger dataset by Chang, R.Y.-W., P.S.K. Liu, W.R. Leaitch and J.P.D. Abbatt, 2007: Comparison Between Measured and Predicted CCN Concentrations in a Semi-Rural Environment: Focus on the Organic Aerosol Fraction, Atmos. Environ., 41, 8172-8182.

Another omission that was corrected (P27651, L18):

"Chang et al. (2007) reached similar conclusions in a 14-day dataset of size-dependent aerosol composition in CCN measurements in a semi-rural environment."

15. Page 27653, Line 1 – The following 2008 study used kappa and aWyoming CCNC: Shantz, N.C., W.R. Leaitch, D. Toom-Sauntry, M. Mozurkewich, and L. Phinney, 2008: The effect of organic compounds on the growth rate of cloud droplets during a marine and a forest field study. Atmos. Chem. Phys., 8, 5869-5887.

We added a reference to this paper in the list of observational studies that use kappa.

16. Page 27653, Lines 16-18 – As you later discuss, the IMPROVE data suggest equal amounts of sulphate and organics, so I would also include a plot with the kappa value between that of sulphate and the organics (e.g. 0.3).

This is a plot (the CCN predicted vs. observed using a higher kappa) that we wish we could make but because of the limitations of the instrument setup that we used it just isn't possible. As written in the text (P27653, L5-15), the PCASP has a lower detection limit of 0.122 micron diameter. If the critical dry diameter for droplet activation is below the PCASP detection limit, then we have incomplete information about the particles that are large enough to activate in that environment. Even the lowest SS setting used in the CCN instrument was above 0.2% and so a low kappa must be used in order to keep the critical diameter above the detection limit of the PCASP. If this were not the

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case we would include a figure with a higher kappa to match the IMPROVE data.

17. Page 27654, Lines 5-7 – Because the Wyoming CCNC is based on volume light scattering, the calibration is also a function of the chemical composition of the particles. If your aerosol is more hygroscopic, the droplets will grow larger and for a single calibration factor a certain concentration of CCN will be indicated. If the particles are less hygroscopic but still large enough to activate then the droplets will grow smaller and for the same number of particles relatively fewer CCN will be indicated. Thus, if your calibration was done with a pure ammonium sulphate aerosol but you were sampling an organic-sulphate mix (as suggested by the IMPROVE data) then a bias towards lower measured CCN could result (e.g Shantz et al., 2008), and that may explain why your closure is better for a kappa of 0.1 rather than 0.3. Also, are the sample volumes of the CCNC and the PCASP both referenced to the same pressure and temperature; I don't think that is discussed?

This is a really great point and it is explained well here and in Shantz et al. (2008). The calibration of the CCNC was done with amm sul and so this low bias could be a result of the different droplet growth factors. This was included in the manuscript (P27655, L5) as the most likely reason that the closure indicated very low hygroscopicity aerosols when other observations in the region indicate otherwise. The sample volumes were referenced to the same temperature and pressure.

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

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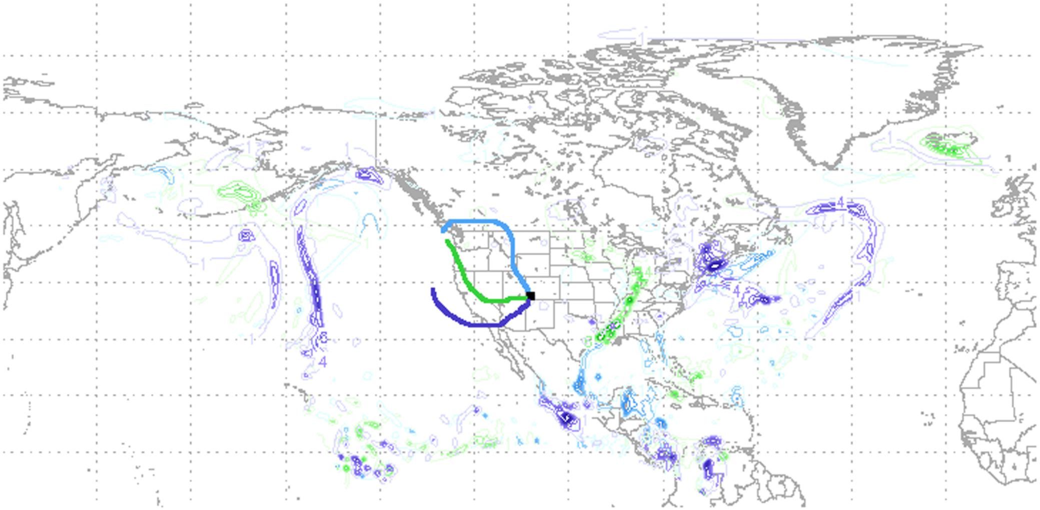

Fig. 1. NARR precipitation and average HYSPLIT back trajectories for each event. The precipitation is color-coded to match up with the corresponding event trajectory. Plotted here are the max Nccn events.

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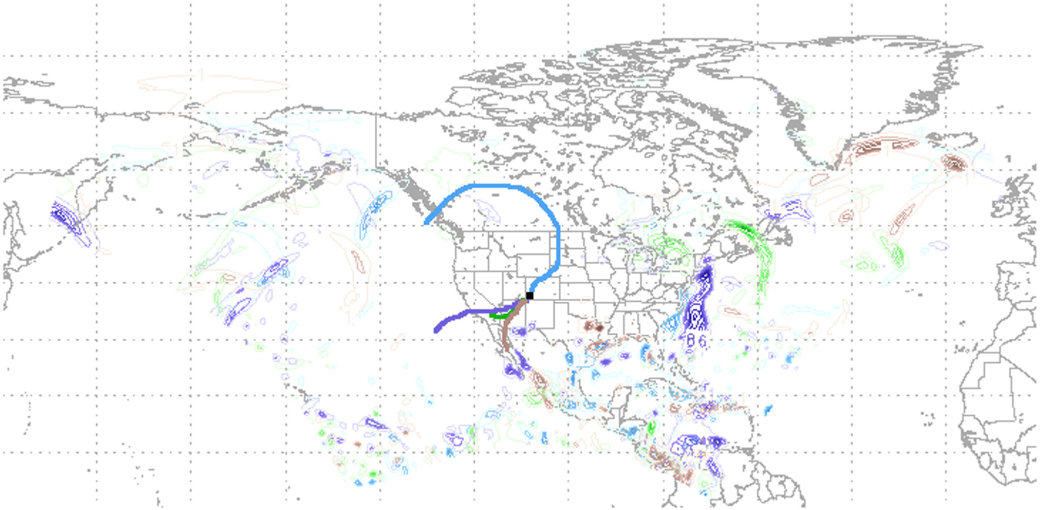

Fig. 2. NARR precipitation and average HYSPLIT back trajectories for each event. The precipitation is color-coded to match up with the corresponding event trajectory. Plotted here are the min Nccn events.