The manuscript presents an analysis of a combination of in-situ measurements, including aerosol chemistry and size distribution, to interpret their relationship with the measured cloud condensation nuclei number concentration. All figures are clear and legible, and the paper is very well written. The manuscript presents a detailed methodology section, an overview section including aerosol composition and CCN number concentration, followed by the main part of the discussion that focuses on predicting aerosol hygroscopicity/CCN number concentrations using a number of different approaches, and validating these approaches against the measured CCNC.

These approaches include the conventional k-Kohler method, using the particle number and size information and a kappa parameter. The second method calculated aerosol hygroscopicity based on aerosol chemistry measurements; then two subsequent 'schemes' provided detailed organic aerosol hygroscopicity. This later method was determined using results from PMF calculations on the ACSM organic aerosol, which separated the organics into three main factors, More Oxidised Oxidised organic aerosol (MO-OOA), less oxidized OOA (LO-OOA), and hydrocarbon OA (HOA). This was followed by another scheme calculating the organic hygroscopicity using f44 (an indicator of aerosol oxidation state) and then finally a method using neural networks was presented. This method included additional variables; Organic fraction (OA/PM1), particle number concentration (N80), and also solar radiation fluxes.

The initial comparisons showed that the hygroscopicity schemes using more detailed information on organic aerosol oxidation state and its hygroscopicity, resulted in much lower aerosol hygroscopicity (and lower variability) than those calculated using K-Kohler theory. These were illustrated using violin plots and later using scatter plots. The scatter plots allowed the assessment of the range of the goodness of fit. Using a proxy for the oxidation state, f44, alone did not result in any improvement and in all cases there was an overprediction of aerosol hygroscopicity during the night and the opposite during the day. The final comparison using neural networks (including 4 variables) worked very well in comparison to CCN values (@ SS of 4%).

General comments

1. It would be very useful to include a figure of the SMPS measurements and eventually to illustrate the contribution of different size modes (N_{ait}, N_{nuc}, N_{accum}) over the period of the study. Using the SMPS we can eventually determine to some extent the mixing state of the aerosol population. One would expect a more externally mixed aerosol population during the time of new particle formation.
2. The work showed that the nighttime aerosol hygroscopicity is underestimated but day time is overestimated. One might assume that nighttime aerosol was more internally mixed and daytime aerosol was more externally mixed based on boundary layer dynamics. Can this be inferred from the SMPS measurements?
3. In Figure 5, the data could also be illustrated by coloring the data points using OA/PM1 or by surface global radiation, or f44. This could provide an introduction into why it would be important to include these variables in the neural network.
4. In the analysis using the neural network, how much weight does each one of the variables carry to assure/maintain the good agreement? Does this provide us with additional information of the chemical/physical processes involved in CCNc prediction?
5. A statement is made that the results between the different hygroscopicity schemes are mostly the same with differences only observed during low hygroscopicity periods (or high HOA

events). It would be helpful to see a time series of the CCN and also of the calculated kappa values.

6. The SMPS and the ACSM are both measuring submicron aerosol. The SMPS is a mobility diameter, the ACSM is an aerodynamic diameter. This should be mentioned or at least convert the SMPS diameter to Aerodynamic when commenting on the comparison of both methods. This is also important for the comment in line 650.

7. *Line 429: The size cut for the CCN measurements can be determined using the aerosol particle loss calculator and information on the aerosol sampling inlets at the site. If the author does not consider that the three instruments (SMPS, ACSM, and CCNC), are measuring the same size range it does not make sense to be compare them, or to use variables from one instrument to predict the measurements from another.*

8. Can these size-cut differences explain the variability of the results, especially during the day when nucleation events occurred?

9. The supersaturations measured in by the CCN range from 0.2 to 0.6, what are the typical supersaturations observed in clouds?

10. Fig. 5 We also see that at low SS (0.2%), NCCN agrees well at low number concentrations and deviates from the line at higher CCN. In previous studies, the impact of Ntotal on NCCN has been shown to have an impact. Do the authors consider that the total NCCN affect the predicted results here?

11. How does the fOA vary over the data points in Fig.5?

Minor comments:

Line: 164: NCCN this is the first use of NCCN, please define.

Aerosol chemistry measurements

What is the impact of the capture vaporizer on the aerosol mass spectra? The capture vaporizer results in a high degree of fragmentation compared to the standard vaporizer. How does this impact the resolved factors in the PMF results?

Looking at the overall aerosol composition, it would be expected to have an acidic aerosol population? Is this the case? Do you expect some different forms of organics to be present?

It is mentioned that the high concentrations of SO_4 measured at the site is from high SO_2? Is SO_2 measured? Is it expected this SO_2 to be transported over long distances or is there local sources. How do these composition compare to other altitude stations.

Being an altitude station, is this station often in cloud? Was the station in cloud during these measurements? Such as around the 6th of June and the 20th of June when the humidity approaches 100%.

Measurements with an AE33 were made. What is the contribution of absorbing aerosols in the other wavelengths compared to those at around 660 nm for BC? Could these increased be a result of other types of aerosols?

In Figure S2: Are error bars available for these plots? Are these differences significant?

Was there a change in wind direction or airmass source during the first and second halves of the field campaign. An analysis of airmass history and back trajectory would be welcome in this work.

Is the site in the free troposphere at night compared to the day time?

In previous work, it was mentioned that NPF is a major source of CCN at this mountain sites. In this work, you also had chemical composition. What were the chemical signatures during these NPF events.

In Figure 3: Having grid lines on the time of day would help to guide the eye of the reader. If this is possible.

Figure 3: Since the magnitude of values between the day and the night are being compared, it would be appreciated to include error bars on these graphs. This would help determine the significance of the differences.

In Figure 2 a time series of aerosol properties is shown which illustrates how the composition changed over time. Were airmass trajectory calculations made for this study?

Line 651: "None size cut" , change to no size cut. Although there is no size cut for this instrument, there is still a size cut implied by the inlets. This can be shown here. In this sentence, a comparison is made with an aerodynamic diameter with a mobility diameter. These are not exactly the same and should be considered.