Response to the reviewer's (Martin Gysel) comments to the manuscript "Relationships between particles, cloud condensation nuclei and cloud droplet activation during the third Pallas Cloud Experiment" (submitted to ACP)

We would like to thank the reviewer for detailed and insightful comments. These are shown below in cursive font. After each comment, our reply is shown in normal font.

Major comments

1) p. 13706, l. 1-4: A positive correlation betweenD50 and CCN(0.4%) is reported here and it is speculated that "This can be feasibly interpreted so that larger numbers of CCN led to decreased activation efficiency due to competition between particles for water vapor during the cloud formation." However, this speculation is in contradiction to the statement made on p. 13703, l.1-3: "No visible correlation can be seen between D50 and CN(>100 nm) which suggests that the observed variation in D50 was not driven by the aerosol number concentrations." Furthermore, the authors essentially acknowledge themselves on p.13706, l. 4-9 that the correlation between D50 and CCN(0.4%) is most likely just a random result caused by poor statistics (just three cloud events): "Interestingly, only weak correlation was found between D50 and CN(>100 nm) while CDNC and CN(>100 nm) were correlated to a significant degree (Sect. 4.1). When limiting the comparison between D50 and CN(>100 nm)for the time periods during which the CCN measurements are available, however, a positive correlation with the coefficient of determination being 0.78 is seen (not illustrated here)." In conclusion, the question whether or not the CCN number concentration has an effect on the D50 has to be addressed in a more stringent way. A good estimate of CCN(0.4%) can be obtained for the whole data set by calculating the critical diameter for CCN activation at SS=0.4% from the time-resolved hygroscopic growth factor measurements (the GF values given in Table 1 indicate that kappa varies between 0.064 and 1.61, resulting in critical activation diameters of 81-110 nm, assuming T=25 °C) and then integrate the number size distributions above this time-resolved critical diameter. If there is no correlation between this estimate of the CCN number concentration and the D50 for the whole data set, then the correlation observed for the limited data set is likely just random. However, statistics would remain limited and therefore the parcel model should be used to systematically quantify whether water vapor competition effects are to be expected in the observed range of CCN number concentrations.

To address this, we have calculated CCN(0.4%) for the cloud cases A and E using the model applied in the CCN closure (described in Section 3.2) rather than relying on the approach proposed by the reviewer. This was done to maintain consistency with the rest of the calculations and also because the model accounts for the mixing state of particles, in contrast to the method described above.

The results of these calculations show that the correlation decreases remarkably when the calculated CCN concentrations for the cases A and E are included in the comparison, the degree of correlation between CCN(0.4%) and D_{50} decreases to 0.03. This confirms that our initial interpretation of the results is not valid. Hence we have rewritten the last paragraph of Section 4.3 to reflect these points.

Further down in the manuscript (p. 13710, l. 1-4) comes a statement that the model indicates effects of the CCN number concentration differences on the resulting supersaturation. The discussion of the influence of CCN number concentration based on different analyses should be connected and consistent.

Both D_{50} and s_{max} are indeed related to the efficiency of the cloud droplet formation among a particle population. This point is now addressed in the manuscript as follows (located at the end of the sixth paragraph of the section):

"It is worth noting that CCN(0.4%) was not correlated with observational D_{50} (Section 4.3) when calculated CCN concentrations were included into the comparison to fill the gaps in the measurements. To compliment this result, we made similar comparison for the model based values of D_{50}. As expected, D_{50} was positively correlated with CCN(0.4%) even though the degree of correlation was rather low, being 0.41 (not shown). Consequently, D_{50} is not as sensitive to the total number of CCN active particles as s_{max} even though D_{50} is also related to the activation efficiency of a particle population."

2) Table 1: The kappa values reported in Table 1 are inconsistent (too small) with the GF values. It seems that a water activity of 0.9 was used to infer the kappa values rather than an RH of 90% (the corresponding water activity will be lower due to the Kelvin effect). For example the kappa value corresponding to GF=1.24 at RH=90% and T=20 °C is ~0.118. The kappa values should be corrected. The difference in kappa values is small, however, it will also systematically affect the hygroscopicity-CCN closure presented in Section 5.

This is indeed the case: the values of kappa were calculated at the water activity of 0.90. However, both the model used in the CCN closure calculations and the cloud parcel model calculate the particle CCN activity correctly by accounting for the temperature-dependent Kelvin effect. On the other hand, the calculations shown in Table 1 were done rather quickly on an Excel spreadsheet as neither of the codes give kappa values as output. Therefore the kappa values were calculated separately.

To conclude with, we re-calculated the kappa values displayed in Table 1 but no other changes were needed.

3) Sect. 5: This section is considered to be rather weak for several reasons:

a) The accuracy of the SMPS is a crucial factor for the result of hygroscopicity CCN closure studies. Was the SMPS compared against an independent measurement? E.g. comparison of the integrated CN number concentration measured by the SMPS compared with a total CN measurement made by a CPC, of course restricted to time periods when only few particles were present in the lower cut-off range of the SMPS (i.e. no nucleation mode present).

The DMPS system on the site is operated according to the GAW protocol (Hatakka et al., 2003) and is compared continuously against a reference DMPS instrument. This is now mentioned in the experimental section.

b) Why is the CCN closure only done for the cloud periods? Instead it should be done for the whole measurement campaign in order to get a more representative picture. Or is it expected that the closure would be systematically different between cloud free and cloudy periods?

The reason is that we consider only the cloudy periods in the current manuscript and an extensive set of CCN calculations, covering the whole campaign, will be presented separately in Jaatinen et al. (2012). This is now mentioned in the end of the first paragraph of Section 4.3.

c) It is speculated that the observed closure bias might be caused by the dependence of kappa on water activity. This might indeed play a role for low SS with high critical diameter (particularly for such low kappa values as reported in this study). However, the biggest closure bias is observed at the highest SS,where the CCN number concentration becomes very insensitive to the kappa value (see e.g. Fig. 8 in Juranyi et al. (2010) for a detailed sensitivity analysis. The key question here is whether the 20% bias are within or outside the experimental uncertainty of the SMPS data.

Thanks for pointing out the importance of experimental uncertainties. We have evaluated the uncertainties in the CCN measurements by taking the standard deviation in the CCN measurements and comparing them to the total CCN number counts. The uncertainties derived this way were, on average, 44, 17, 15, 14 and 11% for supersaturations 0.2, 0.4, 0.6, 0.8 and 1.0%, respectively. These numbers are now reported in the second paragraph of Section 5 and compared to the magnitudes of the biases in the CCN closure.

d) The hygroscopicity-CCN closure results are not at all put in the context of existing literature, e.g. Kammermann et al. (2010) and references therein, Jurányi et al. (2010), Sihto et al. (2012) (closure done for the critical diameter, which makes the comparison a little more difficult) and certainly further recent publications.

The results are now briefly put in the context of previous studies. More detailed discussion will be presented in Jaatinen et al. (2012) because that manuscript covers the whole measurement interval and not only cloudy periods.

e) The HTDMA derived kappa values are not correctly calculated, which affects the closure result (see previous comment made to Table 1).

As noted in our response to the second main comment, the kappa values were calculated correctly in the model calculations.

4) p. 13707, l. 17-18: "The model uses also the Koehler theory which was found to explain the CCN activity of the observed particle quite accurately (Sect. 5)" It is not directly obvious why the hygroscopicity-CCN closure should play a role for the box model. Sect. 3.3 gives the answer: The hygroscopicity of the aerosol was initialized using the HTDMA data. Would it be possible to initialize the aerosol hygroscopicity directly based on the CCN+SMPS data? This approach might result in less accurate description of the hygroscopic growth at subsaturated RH before activation in favor of a more accurate description of the activation behavior of the particles at supersaturated RH, which is of course more important for the cloud simulations. The mixing state information would be lost or could be taken from the HTDMA (taking just the GSD translated to kappa-variability for the HTDMA). Anyway, the influence of the closure bias on the uncertainty of the resulting updraft velocities / cloud peak supersaturations should be quantified (together with other uncertainties such as that of the SMPS number concentration).

First, we have revised the cited sentence to read as follows: “The model uses also the Koehler theory and the critical supersaturations are calculated on the basis of the H-TDMA data (Sec. 3.3). This approach was found to explain the CCN activity of the particles quite accurately (Sect. 5).”

Second, we have actually already done what the reviewer proposed: as discussed in page 13711, lines 1-20, we did increase the particle hygroscopicity so that CCN closure is obtained at 0.4% supersaturation (which is close to estimated supersaturations reached during the cloud events). The results are further discussed in the last paragraph of Section 6.1. Hence we made some changes to the text so that the motivation of the sensitivity study becomes clearer.

5) p. 13708, l. 11 - p. 13709, l. 2: Good reproduction of the measured activation curves by the model is obtained for cloud periods D and E, while substantial differences of the shape and D50 are reported for cloud periods A-C. The authors state that the reason for this remains unknown. One obvious difference in the experimental results is that 100% activation is reached during cloud events D and E, while a stable activation plateau at around 75-90% is reached at diameters >150 nm for cloud events A-C (Why are the activation curves only shown up to 250 nm while the measurement was done up to 500 nm?). Such activation plateaus below unity can be caused by either the presence of an externally mixed non- or less hygroscopic mode or by cloud processes. Mixing state

effects are excluded by the authors, leaving cloud processes as the cause of the activation plateaus below unity. Such activation plateaus can either be caused by entrainment or by evaporation of cloud droplets due to the Bergeron-Wegener-Findeisen process in mixed phase clouds. Such activation plateaus have previously been reported by e.g. Henning et al. (2004) and Verheggen et al. (2007). The parcel model has to be adapted in cases with a non-unity activation plateau (cloud periods A-C) using reasonable assumptions to simulate entrainment or droplet evaporation and using the plateau level as a constraint for the degree of these processes, such that the activation plateau is eventually reproduced by the parcel model. Will the modified simulations change the estimated updraft velocities and peak supersaturations substantially?

First, the activation curves were shown only for the range 50 to 250 nm for the following reasons: 1) it covers the diameter range most relevant to the cloud droplet activation, i.e. the part where the activated fraction increases from zero to unity, and 2) because of the low particle concentrations encountered during the campaign, there were only a few particles sampled per DMPS channel at larger sizes which makes the activation statistics extremely unreliable: estimated errors for the activated fractions above 250 nm were between 35 and 55% on average among the analyzed cloud cases. Also, activated fractions of unity are consistently within experimental uncertainties at this size range. This alone makes it difficult to quantify the role of the discussed processes.

Including entrainment process to the model is also problematic for the following reasons. First, entrainment decreases the activated fraction of particles throughout the particle size range (see e.g. Figure 5 in Noone et al., 1992) and thus adjusting the model to reproduce the "plateau" at larger sizes by including entrainment would imply that activated fractions at lower sizes would not match the observations anymore. Second difficulty stems from estimating the model parameters associated with the rate of entrainment. In the case of cumulus or stratocumulus convection, this would require vertically resolved measurement data on cloud liquid water path (Morales et al., 2011). Due to the nature of the experimental set-up, such data is not available. As the values parameters may vary over a large interval (Barahona and Nenes, 2007), performing simulations while using "educated guesses" for the unknown parameter values is not feasible.

All these factors make quantitative investigation extremely difficult if not impossible. Instead of performing additional simulations, the possible roles of the mentioned processes (entrainment and Bergeron-Wegener-Findeisen) are now discussed on a qualitative level in the third paragraph of section 6.1.

6) Fig. 6: The shape of the measured activation curve is surprisingly well reproduced by the parcel model. The width of the activation curve should be determined by the degree of external mixing of the aerosol as well as the inhomogeneity of the updraft velocity at cloud base. To my understanding the hygroscopic mixing state was considered with the parcel model, while no updraft velocity fluctuations were simulated. Does the agreement between experiment and model imply that the observed width of the activation curve can fully be explained by the external mixing of the aerosol, while updraft velocity fluctuations only had a marginal broadening effect on the activation curve?

Indeed, no updraft fluctuations were simulated as the applied model is zero dimensional. And to answer the question – yes, according to model results, external mixing is sufficient to explain the shape of the activation curves. This agreement should be interpreted with caution, however, because investigating the role of velocity fluctuations would need more information on the origins of the observed clouds and also more detailed modeling tools, i.e. one or two dimensional cloud models. Hence this conclusion is tentative. This is now briefly discussed in section 6.1 (third paragraph).

7) p. 13710, l. 21 - p. 13711, l. 20: It is observed that the CCNC measures higher CCN number concentrations than the observed CDNC at equal supersaturation. Some reasons

for this discrepancy are appropriately discussed. However, there are further caveats:

a) Any observed difference should be put in the context of experimental uncertainties. This is done for the kappa uncertainty. However, for example a difference of 15% in number concentration would fall within experimental uncertainty if the SMPS was undercounting by 10% and the CCNC was over counting by 5%.

We now bring out the experimental uncertainties, estimated as described above (see our response to the comment 3c) and compare them to the magnitude of the differences in the last paragraph of Section 6.1.

b) There is a conceptual problem in the way how the CCNC measurements and the CDNC number concentrations are compared. The Kelvin effect introduces considerable temperature dependence in the CCN activation behavior, even if the kappa value at activation is assumed to be independent of temperature. To give an example, prescribing a kappa value of 0.1 and a supersaturation of 0.4% results in critical activation diameters of 94.8 nm and 105.6 nm at temperatures of 25 °C and 5 °C, respectively. This activation diameter difference roughly corresponds, according to Figs. 8 and 10, to a change in maximal cloud supersaturation from 0.4% to 0.3%. Consequently comparing CCN number concentration measurements made at ~25 °C in the CCNC column directly with CDNC number concentrations of a cloud with a temperature of 5 °C at cloud base is like comparing apples and oranges. However, there is a way to get around this issue: In the first step the CCN number concentrations and SMPS size distributions are used to infer a critical activation diameter corresponding to the supersaturation and temperature in the CCNC. These values allow it then to calculate a kappa value of the aerosol at the activation diameter. This kappa value is in a second step used to calculate the corresponding activation diameter at the cloud base temperature of the parcel model (assuming that kappa is temperature independent and only weakly dependent on particle size). Integrating the CN number size distribution above this corrected activation diameter then provides a corrected CCN number concentration measurement recalculated to cloud base temperature, which can be directly compared with the CDNC values.

First, regarding the sensitivity of s_{max} to kappa values. According to the argument presented above (which, in turn, is based on Figures 8 and 10), decreased particle hygroscopicity leads to a decrease in s_{max}. The conclusion is not physically realistic as decreased particle hygroscopicity leads to decreased competition for water vapour during the cloud formation and hence to larger supersaturations. The flaw in the reasoning lies in using Figure 8 to deduce a relationship between s_{max} and V: Figure 8 shows the s_{max} and V separately for each case. Comparing results between the cases in this manner is not reasonable as s_{max} is affected also by the CCN concentrations which vary between the events.

Nevertheless, the reviewer is correct in pointing out that the temperature influences CCN concentrations. Instead of relying on the approach presented above, we used the model described in Section 3.2 to maintain consistency with the rest of the calculations. We did recalculate the CCN concentrations at the cloud base temperature (which was assumed to be the same as those measured in the station) and compared them with the CCN calculations done at 25 °C. On average, the CCN concentrations decreased by 13, 8, 9, 10 and 9% at supersaturations 0.2, 0.4, 0.6, 0.8 and 1.0%, respectively. Assuming that the CCN activity of the measured particles would display similar temperature dependence, the temperature difference is not alone enough to account for the differences between measured CCN concentrations and calculated CDNC at similar supersaturations. As we will now discuss in the manuscript, however, this effect together with experimental uncertainties could explain most of the discrepancies.

The additional calculations are now included in a table (we made a separate table showing experimental and calculated CCN concentrations) and the effect of temperature is discussed in the last paragraph of Section 6.1.

8) p. 13714, l. 19-23: The authors conclude with: "It should be noted, however, that the current study is based on a rather short intensive campaign where the range of atmospheric conditions encountered was limited. Therefore long term simultaneous measurements of aerosols, CCN and cloud droplet activation are desirable to investigate how the results obtained here compare to larger data sets containing results from different seasons and air mass types." I agree that only limited data are available for the variability of updraft velocity and there is not much that can be done about this without massive additional experimental effort. However, I am sure that much more data are available about the variability and mean values of aerosol number size distribution and aerosol hygroscopicity, possibly from previous campaigns. These measurements don't have to be acquired concurrently with CDNC measurements (a reasonable range of updraft velocities can just be taken from this study) and possibly not even within clouds (unless aerosol properties were substantially different during cloud periods than outside cloud periods). Larger and more representative data sets could therefore be considered for the sensitivity analyses made in Sect. 6.2.

We thank reviewer for this suggestion. Indeed, our plan is to consider larger data sets including CCN and hygroscopicity measurements as a part of a future study. The current manuscript is focused on a single campaign and including larger set of data would make need dedicated effort. Besides, it would make the manuscript incoherent and excessively lenghty. Hence we settle on stating our future goals at the end of the summary and conclusion section.

Minor comments

9) p. 13693, l. 28-29: Further closure/sensitivity studies of this nature are e.g: Kammermann et al., 2010 and Jurányi et al., 2010

These studies are now mentioned.

10) p. 13696, l. 16-20: Average meteorological conditions would be at least as interesting for the cloud periods only, as the majority of data is reported for cloud periods.

Table 1 gives temperature and visibility during the events and air mass trajectories are now shown for each event. This should be sufficient information.

11) p. 13697, l. 22: Briefly explain how the CCN was calibrated?

CCN was calibrated on-site using a DMA (short HAUKE type) coupled with a CPC TSI 3010 and Aerosol Generator ATM 226 (Topas GmbH, Germany) with ammonium sulfate solution in a temperature difference range of 2 to 16 Celsius, thus covering SS range from about 0.1 to 1 %. This is now mentioned.

12) p. 13697, l. 25: Five minutes may not be enough time for the CCNC to fully stabilize when the applied SS is changed from 1% to 0.2%. The DMT CCN indicates "stabilized" temperatures much earlier than this is truly the case. Please confirm that stabilization of the CCN instrument was carefully assured.

Yes, we agree with the reviewer. In our SS cycles (0.2 to 1%) one blind 5 minutes cycle at SS= 0.2% after SS=1% is used to fully stabilize the column temperatures. This blind cycle is not used for data analysis.

13) p. 13698, l.3-5: a) How was the ambient RH measured? Measurement of RH close to 100% RH may be difficult depending on the method. On the other hand, using a dew point measurement behind the total inlet and a reliable ambient temperature measurement can provide a reliable measurement of the total cloud water content.

b) How was the visibility determined, particularly during night time?

Visibility was measured with Vaisala FD12P weather sensor both day and night time, and the ambient RH was measured with Vaisala HUMICAP sensor. The instruments are now measured in the manuscript (Section 2.1).

14) p. 13699, l. 20: Here it is described how the interpolation in size space is handled. What about the interpolation in time? Is the size dependence fitted for each full cycle of HTDMA data?

Yes, this is correct, and is now mentioned in the manuscript.

15) p. 13700, l. 12-15: Fitting the measured growth factor distribution with a lognormal function before determining the activated fraction is an unnecessary approximation step which potentially introduces errors. The activated fraction can directly be calculated from the measured growth factor distribution without any approximation (except for interpolation in size and time space of course), as shown in detail by Kammermann et al., 2010. The errors introduced by the lognormal approximation are likely very small, however, it is often simpler to make an accurate calculation without approximations rather than arguing that certain approximations don't cause substantial bias.

The applied model requires that the cumulative distributions of hygroscopic growth factors are described with an analytical function (see Section 2.1 in Anttila et al., 2009). While the following advantages are not relevant to this study, this assumption allows for deriving CCN parameterizations for large scale models and also makes certain kind of sensitivity studies easy to perform (see Anttila et al., 2009, section 4.4). Consequently, getting rid of the assumption would require implementation of a different model which is beyond the scope of the current study.

16) p. 13701, l. 14: How sensitive are the model results to the assumed mass accommodation coefficient of water? Is it possible to give some kind of a limit below/above which the model results are sensitive/insensitive to changes in the mass accommodation coefficient?

The estimated updraft velocities display a gradual sensitivity to the value of mass accommodation coefficient (alpha) so that they decrease by around 25% when alpha is decreased to its lowest recommended value which is 0.04 (Laaksonen et al., 2005). We now address the issue in the beginning of Section 6.

17) p. 13702, l. 19: Please explain in the experimental section that the CDNC is indirectly obtained from the difference of the particle number concentrations behind the total and interstitial aerosol inlets. Henning et al. (2002) nicely showed for liquid clouds that this indirect approach provides reliable values of the CDNC.

We have now briefly described the procedure and cited previous studies where the method was applied.

18) p. 13702, l. 24: It would be nice to compare the D50 values observed in this study with D50 values from other sites (e.g. Henning et al., 2002).

The D_{50} values are now compared with results from other sites.

19) p. 13703, l. 25: I guess that the GF values are interpolated in diameter space and averaged over the duration of the cloud events.

This is correct, and the issue is now clarified in this part of the manuscript.

20) p. 13704, l. 16: It might be worth repeating here that the Pallas site has a strong boreal influence.

We do not wish to expand the discussion to cover the origins of the organic aerosol matter, but will present a detailed discussion in Jaatinen et al (2012). Hence no changes were made.

21) p. 13705, l. 10: It would not be out of scope to put the observed kappa values briefly into the context of results reported from the boreal site Hyytiälä (Cerully et al., 2011; Sihto et al., 2012).

The results from these studies are now mentioned in the second paragraph of Section 4.2.

22) p. 13712, l. 20-21: Sensitivity analysis using the parcel model: "...For the cloud events B and C, however, the modeled values of CDNC showed largest sensitivity to the particle hygroscopicity..." This statement is potentially misleading as it seems to imply that cloud events B and C are more sensitive to aerosol hygroscopicity than to aerosol size distribution. However, this result possibly just reflects that for these two cloud events the CN size distribution in the CCN active size range was close to the averaged size distribution, while the aerosol hygroscopicity was clearly lower than the averaged aerosol hygroscopicity.

This is a good point. By looking at Table 1, it can be seen that for events B and C, the case averaged values of CN(>100 nm) did not deviate much from the overall average value of CN(>100nm) (which was 103 cm^{-3}). On the other hand, Table 1 also shows that the particle hygroscopicity was lower compared to the overall average value, which was 1.21 for 100 nm particles, during the events B and C. These two factors together explain the result, and the issue is now brought out in the manuscript.

23) Fig. 5: It should be emphasized in the figure caption that this excellent agreement is forced by varying the updraft velocity in the model until agreement is achieved. This figure is all about showing that the chosen resolution of 0.05 m/s in updraft velocity steps is sufficient to reproduce the CDNC reasonably close. Actually, iteratively approaching the best fit updraft velocity with e.g. a simple bisection method rather than using a fixed updraft velocity grid would have provided perfect agreement between experiment and model with fewer model runs, thereby making this "verification"-figure obsolete.

To be exact, perfect agreement cannot be reached because the particles are discretized into a finite number of bins in the model. However, on the basis of this comment and a comment from the other reviewer, we have decided to omit the figure and instead report the error numbers.

24) Fig. 10: The data points in this figure could be colored by e.g. the CDNC or CN(>100 nm) in order to see whether the deviations from the fit line are related to the availability of CCN.

We have modified the figure so that the data points are colored according to the cloud event as requested by the other reviewer. While this not exactly what was suggested, the reader can find the average CCN concentrations from Table 1 for comparison purposes.

Technical corrections:

25) p. 13699, l. 19-20: Suggestion: "Accordingly the experimentally determined size dependence of the hygroscopicity parameters ..."

This was corrected to read as follows: “Accordingly the hygroscopic parameters….”

26) p. 13703, l. 9: The reference should be to Fig.2 instead of Fig. 1.

The reference is now given to the correct figure.

27) *p. 13704, l. 19: Add the Jaatinen et al. (2012,in preparation) paper to the reference list (with the tentative title and author list).*

This is now done.

28) p. 13705, l. 15: Do you refer to Fig. 2 or Fig.3?

Figure 2 is correct.

29) p. 13713, l. 27: Should read: "The second part of the modeling ..."

The sentence has been corrected.

30) Table 1: Reporting GF values without specifying the corresponding RH is useless.

The RH is now specified.

References

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