## *Reply to interactive comment from Referee #1 on*: 'Technical Note: Using DEG CPCs at upper tropospheric temperatures' by D. Wimmer et al.

We thank the referee for her/his review and comments on the draft that improved the manuscript considerably. We will address the comments pointby-point in the sections below.

**Comment 1**: The manuscript states that "There is a clear shift in the cut-off diameter towards larger sizes at lower temperatures". One may argue that clear evidence to reach this statement is missing, i.e., the counting efficiency of freshly formed particles from the CLOUD chamber at room temperature. Comparing to the counting efficiency of ammonium sulfate at room temperature was unconvincing to draw this statement since particle composition strongly affects the counting efficiency.

**Reply**: We agree with the reviewer's comment. The comparison should be done for the same chemical system also at room temperature. To confirm that we can state that there is a shift in the cut-off size and that the lab measurements can be used as the reference for room temperature we analyzed an experiment done at 294K in the CLOUD chamber. We used CLOUD5 data for the binary system to estimate the cut-off of the DEG CPCs for room temperature conditions. The Figure shows the ratio between the normalized 60 second averaged concentrations PSM/DEG-CPC1 and PSM/DEG-CPC2 for one specific run in CLOUD (644.01) with chamber temperature 294K and RH 20%. This result is comparable to the results from the laboratory calibrations at room temperature.

The cut-offs for both DEG CPCs in these conditions are around 1nm and therefore considerably smaller than at colder temperatures, but we have to keep in mind that we know that for runs with temperatures above 278K, there is always some contaminant ammonia present in the clusters. Nevertheless, we see this as a sufficient confirmation for our original statement.



The Figure shows the ratio between normalized DEG-CPC/PSM as function of particle size as evolving during the run, starting at 0.5 nm. The resulting cut-offs are ~1nm.

We added the following sentence to the manuscript:

p. 10, 5-8: Nevertheless there is a clear shift in the cut-off diameter towards larger sizes at lower temperatures. Using CLOUD run data at higher temperatures for analysing the cut-offs of the DEG-CPCs at room temperature (293 K) showed comparable results to the laboratory calibration measurements (d50 ~1nm).

**Comment 2**: The statement that growth rates calculated from three instruments converge at high temperature is unconvincing from Figure 4. In the current linear scale, the low values in growth rates at higher temperatures may disguise their differences.

**Reply:** The Figure 4 has been changed to a logarithmic scale on the y-axis. The Referee is right that the points do not converge, in fact the difference between the points is very similar for all temperatures. The GRs calculated from the NAIS agree within a factor of 1.7 at +5C with the results from SAWNUC, whereas the corresponding factor is 5 at -50C.

**Comment 3**: The authors stated that the problem of low signal to noise ratio is reduced by using PSM instead of aerosol electrometer as a reference method. Please discuss the uncertainties and problems introduced by making this switch. For instance, PSM itself may exhibit strong temperature effect in its counting efficiency.

**Reply**: It is true that the PSM as the reference is not an optimal solution either. In order to avoid this we tried to minimize the effect of the temperature

on the counting efficiency of the PSM by adjusting the settings at each chamber temperature so that the background concentration of the PSM was the same, which indicates a similar cut-off diameter. We added some text to the manuscript to explain this issue in more detail, as follows:

P. 6, I. 16-25: We assume that the PSM behaves similarly to the laminar flow DEG CPC regarding the ambient temperature. To ensure that the cut-off diameter of the PSM stays the same despite varying experimental temperatures, the saturator temperature of the PSM was adjusted for each temperature in the CLOUD chamber. By keeping the homogeneous background in the PSM constant, the assumption is made that the supersaturation within the instrument stays consistent. More detailed discussion about the influence of the temperature settings on the instrument behaviour when using an ultrafine DEG-based CPC can be found in e.g. Jiang et al 2011a, Kangasluoma et al, 2015}. These assumptions have not been quantified and certainly add some uncertainty to the results. It would be highly interesting to investigate the effect of the ambient temperature on ultrafine CPCs further, but this is not in the scope of this technical note. With this setup, the problem with a too low signal to noise ratio could be reduced and in frame of the measurements presented here, were the best results we could achieve.

**Comment 4:** Though this manuscript refers to Wimmer et al (2013) for details about DEG UCPCs, some key information should be provided to make the manuscript self- sustained, e.g., the temperatures of the saturator and condenser for both CPC 1 and CPC 2. Which CPC was used to obtain the results in Figure 3? Was an aerosol electrometer or PSM used as the reference to obtain the black curve in Figure 3? If it is an aerosol electrometer, please discuss the suitability in comparing them with data using PSM as the reference, i.e., blue, green, and red curves in Figure 3.

## **Reply**: we have addressed these issues in the manuscript as follows

p. 5, li. 10-17: The sub-5nm CPCs that were used for measuring the total particle concentrations and calculating nucleation rates were a Particle Size Magnifier (PSM; Airmodus A09), a butanol based laminar flow CPC (3776; TSI Inc.) and two DEG based laminar flow CPCs. Details about the setup and performance of the DEG CPCs can be found in (Wimmer et al., 2013). In short, the DEG-CPC1 was set to a saturator temperature of 325K and an inlet flow rate of 1.5 lpm, whereas the DEG-CPC 2 has a saturator temperature of 328K and an inlet flow rate of 1.8 lpm. The condenser temperatures are 283K in both cases.

In the Figure caption of Figure 3, p.22: Black curve shows laboratory calibrations (Wimmer et al, 2013), using negatively charged ammonium sulfate clusters and a Faraday Cup Electrometer as reference. All curves were determined for the DEG-CPC1, which has a saturator temperature of 52°C and inlet flow rate of 1.5 lpm.

In section: Upper tropospheric temperature calibration results, p. 10, I., 5-8:

Nevertheless there is a clear shift in the cut-off diameter towards larger sizes at lower temperatures. Using CLOUD run data at higher temperatures for analysing the cut-offs of the DEG-CPCs at room temperature (295 K) showed comparable results to the laboratory calibration measurements ( $d_{50}$ ~1 nm).

**Comment 5**: In Figure 3, two different data points were given for some particle sizes. Please explain their differences and which data points were used for the regression.

**Reply:** The calibration measurements using the CLOUD chamber as aerosol generation unit were done by doing several consecutive runs at the same settings. For the results at -50.5°C and -26.9°C one run was used, whereas for the results at -65.9 two sets of measurements were used, which results in different data points at the same sizes. For the fit function, both sets of data points were used.

**Comment 6:** For different CLOUD chamber conditions, temperatures at the inlets of DEG UCPC and PSM should be reported. Otherwise, the findings of this study cannot be readily evaluated or compared by future studies.

**Reply:** There were no temperature measurements at the inlets of the CPCs, nevertheless the chamber temperatures reported in the manuscript were measured at the same height as the sampling lines attached to the CLOUD chamber.

**Comment 7**: Please report whether UCPC and PSM saturator and condenser temperatures are affected by the CLOUD chamber conditions. In addition to reporting the counting efficiency (as given in Figure 3), these temperature information will help to better understand the saturation profile in the condenser and particle activation.

**Reply**: the temperatures both in the PSM and UCPC were monitored all the time and no significant changes could be observed. The Figure below shows the measured saturator and condenser temperature as a function of the chamber temperature for the DEG stage of the UCPC.



The Figure shows the chamber temperature for the full CLOUD5 campaign on the x-axis (in centigrades). Due to a failure of the cooling system of the chamber, runs at uncontrolled, but monitored temperatures were also performed. The condenser temperature of the DEG stage (blue) and the saturator temperature of the DEG stage (red) on the y-axis are shown.

## **Comment 8:** In Figure 2, the two grey lines are not at the half maximum as stated in the manuscript.

**Reply**: we made the point clearer in the manuscript. The tolerance for the half maximum was between 50% and 60%. For the example Figure, the 60% point was chosen. The raw data from the DEG CPCs was averaged using a running average of 5 seconds. That helps to reduce some of the scatter of the raw signal. Nevertheless there is still scatter in the signal from the CPCs, so the 50% point might not be exactly defined. To account for that, we chose to allow for some range when finding the 50% points.

The according text in the revised manuscript:

p. 8, I. 5-10: The raw signal from the CPCs was smoothed using a running average of 5 seconds. Next, the times when the concentrations reached half of the maximum concentrations were determined. The half maximum is defined by the range between 50 and 60% of the maximum concentration as even the smoothed signal still shows fluctuations. The time difference between the half maxima of the two DEG CPCs was then used for determining the growth rates.

**Comment 9:** When discussing CPCs for sub 2 nm particle detection, lida et al (2009) should be cited. After examining more than 800 hundreds working fluids, this reference suggests a few suitable working fluids for sub 2 nm

measurement (including diethylene glycol which has been used by following studies). It also experimentally demonstrates their capability in detecting sub 2 nm particles.

**Reply:** The theoretical analysis presented in lida et al. (2009) provides crucial information on DEG as the working fluid. Leaving this out was an unfortunate omission and we added a citation in the revised manuscript, as follows:

P. 3, I., 4-6: The work presented here, focuses on ultrafince CPCs using diethylene glycol (DEG) as condensing liquid, which has been tested in a study by (lida et al, 2009) and proven to be suited for activation of sub-3 nm aerosol particles.

Comment 10: Line 5 in page 12800, it should be "Outdoor".

**<u>Reply:</u>** In normal typography yes, but The CLOUD-project acronym comes from Cosmics Leaving OUtdoor Droplets

**Comment 11**: Line 28 in page 12802, please replace "make-up flow" with "transport flow". In addition, please add 2.5 lpm dilution flowrate in Figure 1.

**Reply**: corrected in the revised manuscript. The dilution flowrate (2.7 lpm) was added in Figure 1 of the revised manuscript.

**Comment 12**: Line 7 in page 12804, the reference format is incorrect.

**Reply:** corrected in the revised manuscript

Replies to the *Interactive comment of Referee #2 on* "Technical Note: Using DEG CPCs at upper tropospheric temperatures" by D. Wimmer et al.

We thank the Referee for her/his comments and constructive suggestions and we will answer those in the following.

**Comment 1**. P. 12799, I. 1 - 2: The definition of cut-off diameter should be clarified o "counted with 50% efficiency" as opposed to "counted with at least 50% efficiency". With the original definition, a particle diameter with 100% detection efficiency would be considered as a cut-off diameter.

**Reply**: we changed the description in the revised manuscript as follows: P. 2, I. 23-25 The parameter used to describe the performance of a CPC is the cutoff diameter, which is defined as the diameter at which the particles are counted with 50% efficiency.

**Comment 2**. P. 12799, I. 3: While (Kulmala, Mordas et al. 2007) does describe the use of CPCs with different cut-sizes, it makes no mention of using those CPCs for the measurement of growth rates, but rather for inferring particle composition. Please find a different reference.

**Reply**: references to Riipinen et al, 2007, Riccobono et al., 2012 and Lehtipalo et al., 2014 were added to the revised manuscript.

**Comment 3**. P. 12799, I. 5 - 6: Please include a reference to the work of (lida, Stolzenburg et al. 2009).

**Reply:** This was an unfortunate omission and the reference to the work by lida et al has been included in the revised manuscript as follows:

P. 3, I., 4-6: The work presented here, focuses on ultrafince CPCs using diethylene glycol (DEG) as condensing liquid, which has been tested in a study by (lida et al, 2009) and proven to be suited for activation of sub-3 nm aerosol particles.

**Comment 4.** P. 12799, I. 10: The publication year for (Kangasluoma, Kuang et al. 2014) should be 2014.

**Reply:** the publication year was corrected in the revised manuscript

**Comment 5**. P. 12799, I. 10: Please include references to the work of (Winkler, Steiner et al. 2008) and (Sipilä, Lehtipalo et al. 2009).

**Reply:** The suggested references were included in the revised manuscript

**Comment 6.** P. 12799, I. 9: This is more of a general comment in that while this study does present interesting results for the DEG CPC detection efficiency at UT (upper troposphere) relevant temperatures, I suspect that the sample pressure is at ambient pressure, which is not necessarily relevant to the UT. Please clarify this in the text that the DEG CPC characterization does not include calibration at reduced pressures characteristic of the UT (which can have substantial impacts on particle activation through their effect on heat and mass transfer inside the condenser).

**Reply:** The referee is correct. The measurements are done close to ambient pressure at ground level (CERN, Geneva, ~400 m above sea level). In detail, the measurements presented in the manuscript were performed at 4 mbar over-pressure. To avoid accidental contaminations, the CLOUD chamber is operated at slight over-pressure during the nucleation experiments. We included a more detailed description in the revised manuscript as follows:

P. 3, I. 28 – P. 4, I. 6: The aim of this study is to investigate the possibility to use CPCs at temperatures, which can be encountered in the upper troposphere (e.g. during flight measurements). Note that the experiments presented here were carried out at ambient pressure plus 4-5 mbar over-pressure. Therefore, influences of operating the CPCs at UT pressures on the cut-offs and other performance characteristics of the CPCs were not investigated in our study. The chamber is operated at this slight over-pressure to avoid accidental contamination. We studied how the counting efficiency curve of a laminar flow DEG CPC changes when varying the temperature and how the sub-3 nm growth rates behave at low temperatures by using different instruments.

In section: The CLOUD facility: P. 5, I. 3-6: The chamber is operated under over-pressure of 4-5 mbar. Under-pressure is to be avoided, as ambient air, DEG or butanol vapors from the instruments could leak back into the chamber, for example during a pump failure. To guarantee the same temperature in the chamber and in the sampling lines, the sampling lines were thermally coupled to the chamber, using copper.

**Comment 7**. P. 12799, I. 16 - 18: Since this study presents a calibration of DEG CPCs at low sampling temperatures, it would be very helpful/instructive to the reader to present a theoretical calculation/prediction for the effect of low sampling temperatures on the resulting super-saturation profile of the instrument condenser.

**Reply:** The modeling results using COMSOL 5.0 multiphysics show the exact same temperature at the beginning of the capillary. The results were achieved assuming a constant flow of 0.3 Lpm through a tube with a radius of 0.125 cm and a total length of 22.2 cm. The total length was split into three temperature regions. The inlet part with a length of 2.7 cm was set to 208.15K for the low

temperature run and 293.15K for the high temperature run. The part of the inlet, which is inside the CPC was assumed to be at 303.15K, which is the temperature inside the CPC cabin as measured. The part of the inlet, which is in the heated saturator region was assumed to be 2.2 cm long and at 328.15K. The modelling results show the exact same temperature at the beginning of the aerosol capillary. Therefore we can conclude that the CPC saturation ratio profile, is not influenced by the ambient temperature.

We clariefied that in the revised manuscript as follows: P. 10, I. 20-27: Modelling results, using COMSOL 5.0 multiphysics model show that assuming a constant flow of 0.3 lpm from the inlet of the CPC until the beginning of the capillary, the final temperature of the flow is 326 K for an inlet temperature of 208.15 K and the same for an inlet temperature of 298.15 K. The dimensions that were used for the modelling are: 2.2 cm inlet at ambient temperature, 17.3 cm inside the CPC cabin, assuming at temperature of 303.15 K and 2.7 cm inside the saturated region, assuming a temperature of 328.15 K. These results suggest that the ambient temperature does not influence the profile of the saturation ratio in the condenser of the CPC.

**Comment 8**. P. 12799, I. 25 - 26: It is not entirely accurate to say that the ion spectrometer is a temperature-independent measurement method, as the air ion spectrometer classifies aerosol based on their electrical mobility in air, which is a function of air temperature. Please re-phrase.

**Reply:** The part in the manuscript stating the NAIS measurement is temperature independent was re-phrased in the revised manuscript as follows:

P. 3, I. 23-26: The comparison with an ion spectrometer gives important additional information, as the effects of the experimental temperature on the particle detection method, such as volumetric flow rate and mobility to diameter conversion can be accounted for.

**Comment 9**. P. 12799, I. 30: Without also a calibration of the DEG CPC at pressures relevant to the UT, the presented calibration results should not be used to interpret ambient UT data.

**Reply:** we re-phrased the sentence in the revised manuscript as follows:

P. 3, I. 28 – P4, I. 4: The aim of this study is to investigate the possibility to use CPCs at temperatures, which can be encountered in the upper troposphere (e.g. during flight measurements). Note that the experiments presented here were carried out at ambient pressure plus 4-5 mbar over-pressure. Therefore, influences of operating the CPCs at UT pressures on the cut-offs and other performance characteristics of the CPCs were not investigated in our study. The chamber is operated at this slight over-pressure to avoid accidental contamination. We studied how the counting efficiency curve of a laminar flow DEG CPC changes when varying the temperature and

how the sub-3 nm growth rates behave at low temperatures by using different instruments.

**Comment 10**. P. 12801, I. 12: Please specify the manufacturer and model for both the "standard nano-DMA" and the "high-resolution DMA" as you have also done for the CPCs mentioned in the same section.

**Reply**: this was addressed in the revised manuscript as follows:

P. 5, I. 17-23: It should be noted that the DMA that was used for size selecting the particles for the calibration measurements shown here was a commercial nano-DMA (Grimm Aerosol Technik, S-DMA, 55-100, whereas for the calibration measurements at room temperature, a high resolution DMA (Hermann et al, 2000) was used, which means that the calibrations in the laboratory have a better size resolution. A comparison of the performance of the different DMAs can be found in Jiang et al, 2011.

**Comment 11**. P. 12802, I. 5 - 6: Since you mentioned earlier that no aerosol neutralizer was used prior to mobility classification, it is not accurate to state that particle concentrations are low due to "the low charging probability", since the charged particles could have been formed through ion-induced pathways rather than from ambient charging of neutral particles. Please clarify.

**Reply:** True, this was a mistake. We clarified the statement as follows:

P. 6, I.. 9-12: The particle concentrations after the size selection were low due to many factors, including low ion concentrations (about 400/cm<sup>3</sup> (Kirkby et al, 2011)), fast particle growth (i.e. particles grow out of the desired size range quickly) and internal losses in the DMA.

**Comment 12**. P. 12802, I. 8 – 13: The author's justification for use of a PSM as the reference counter rests on the stated assumption that the PSM "behaves similarly to the laminar flow DEG CPC regarding the ambient temperature." It is reasonable to assume that the PSM will still have a lower cut size compared to the DEG CPC at lower ambient temperatures. However, if the author states that the PSM is assumed to behave similarly to the DEG CPC in response to ambient temperature, then it follows that the PSM cut size is also dependent on the ambient temperature as it is for the DEG CPC cut size. If the PSM cut size is itself dependent on the ambient temperatures is not justified. In light of this, the author should provide justification that the PSM cut-size does not change with ambient temperature. Additionally, since a PSM is used as the reference counter in this study, the term "detection efficiency" should be defined in the text/figures/figure captions as "detection efficiency relative to a

## PSM".

**Reply:** yes, it can be assumed that the PSM behaves similarly as the laminar flow CPC with respect to temperature as the method of activating the particles is very similar as in the DEG UCPC. The settings of the PSM were adjusted to the same background for each chamber temperature, which indicates similar cut size. We clarified this point in the revised manuscript. Also the definition was changed according to the suggestion of the referee. Nevertheless we are well aware that the effect of the temperature on the PSM still needs to be kept in mind when looking at the results. On the other hand that was the best option we had for performing the measurements presented here. This issue definitely needs some deeper investigation, which is not in the scope of this technical note.

The additional text in the revised manuscript: P. 6, I. 16-27: We assume that the PSM behaves similarly to the laminar flow DEG CPC regarding the ambient temperature. To ensure that the cut-off diameter of the PSM stays the same despite varying experimental temperatures, the saturator temperature of the PSM was adjusted for each temperature in the CLOUD chamber. By keeping the homogeneous background in the PSM constant, the assumption is made that the supersaturation within the instrument stays consistent. More detailed discussion about the influence of the temperature settings on the instrument behaviour when using an ultrafine DEG-based CPC can be found in e.g. Jiang et al, 2011a, Kangasluoma et al, 2015. These assumptions have not been quantified and certainly add some uncertrainty to the results. It would be highly interesting to investigate the effect of the ambient temperature on ultrafine CPCs further, but this is not in the scope of this technical note. With this setup, the problem with a too low signal to noise ratio could be reduced and in frame of the measurements presented here, were the best results we could achieve.

The term detection efficiency was changed to detection efficiency relative to a PSM in the text.

**Comment 13.** P. 12802, I. 16 - 21: Is the UV turned off after the particles "grow for a couple of minutes"? If the "UV is switched on again to trigger new growth", then it implies the UV was off before then. Please make this clear in the text.

**Reply:** the text was clarified in the revised manuscript as follows:

P. 7, I. 3- 6: Slow enough growth was achieved by periodically switching on and off the UV lights at very low intensities for a few minutes. As soon as the particles grew to sizes bigger than 10 nm, the UV was switched on again to

trigger new growth of small particles.

**Comment 14.** P. 12802, I. 21 - 24: Please provide justification/evidence that the nano-DMA mobility classification at such low temperatures is calibrated (with ion mobility standards for instance).

**Reply**: the inversion from the voltage applied to the center electrode to the diameters was corrected for the according temperatures during the experiments. Also the volumetric flow rates were corrected for the different temperatures.

**Comment 15.** P. 12802, I. 29 – 30: Based on the setup in Figure 1, the 10 lpm flow stream should be described as transport flow, not "makeup flow".

**Reply:** We are sorry for this mistake in the terminology. It is corrected in the revised manuscript.

**Comment 16.** P. 12803, I. 4: Based on the finite width of the nano-DMA transfer function (as described in (Jiang, Attoui et al. 2011), please propagate the resulting spread in mobility diameter for the classified aerosol and include a quantitative description of that spread either in the text or in the data points of Figure 3. The way detection efficiency is presented in Figure 3 suggests that the mobility classified aerosol is mono-disperse, which is not accurate. Also, have the detection efficiency values in Figure 3 been corrected for the finite width of the nano-DMA transfer function as described in the appendix of (lida, Stolzenburg et al. 2009)?

**Reply:** Additional description to account for the finite width of the nano-DMA, following the paper of Jiang et al, 2011 was added to the revised manuscript, as follows:

In Figure caption of Figure 3, P. 22: The error, resulting from the finite width of the transfer function of the nano-DMA, used for the low temperature calibration measurements, is according to Jiang et al, 2011a, 0.185 at 1.47 nm, 0.158 at 1.78 nm and 0.14 at 3 nm."

Equation B6 in the appendix of Iida et al, 2009 was used to account for the diffusion losses after the DMA. We calculated the diffusion losses from after the DMA until the particles reach the inlet of the CPC/FCE. This is justified as the results in the appendix show, that the difference between the formal inversion using eq. B1 and the approximation shown in eq. B6 is negligible. Additionally we assume that the transfer function and the size distribution after the classification are the same for the PSM and the UCPC.

**Comment 17**. P. 12803, I. 6: It appears that two kinds of PSMs are used in this study, one mode where the PSM is used as a total counter (detection

efficiency), and one mode where the PSM is used to determine growth rates (is this a scanning PSM or a total counter PSM used in conjunction with another CPC?). Please make this distinction clear in the text.

**Reply:** Yes, the referee is right. For the calibration measurements, one PSM was used as a total counter, whereas for the calculation of the growth rates, two PSMs with different cut sizes in conjunction with other CPCs were used. We clarified that in the revised manuscript as follows:

In section: Calibrating CPCs using the CLOUD chamber as an aerosol source, P. 6, I. 12-13: This resulted in an unacceptably high noise to signal ratio and therefore it was decided to use a PSM at a fixed cut size as the reference instrument instead of using an electrometer.

In section: Method for determining growth rates from the DEG CPCs, P. 7, I. 22-26: To determine whether the shift in the cut-off curves influences the scientific outcome of the measurements with the laminar flow DEG CPCs significantly, the growth rates have been determined and compared to other instruments (NAIS, Mirme et al, 2013) and PSM). The growth rates from the PSM were determined by using two PSMs at different cut sizes together with the other ultrafine CPCs.

**Comment 18**. P. 12803, I. 15: Based on the text "the method for calculating the growth rates from these instruments had to be determined", it seems that the author is presenting what is described as new method for determining growth rates. This is a bit outside of the scope of the study as described in the abstract. If no new methods were developed (determined), please amend the text.

**Reply**: the text has been changed according to the suggestion of the referee as follows:

P. 8, I. 4: The sentence before "An example illustrating the chosen method is shown in Figure 2. " has been deleted in the revised manuscript.

**Comment 19.** P. 12803, I. 19 - 21: The sentence construction is unclear for this sentence: "Based on the rise times of the different CPCs used here (PSM d50 = 1.1 nm, TSI 3776 d50 = 2.9 nm), the difference in the cut-off diameter of the two DEG CPCs was estimated to be 0.3 nm." How does "the difference in the cut-off diameter of the DEG CPCs was estimated to be 0.3 nm" follow directly from "Based on the rise times of the difference CPCs used here." The two statements are entirely independent of each other. Please re-phrase for clarity.

**Reply:** Correct, the reasoning was confusing. The point is that the difference in cut-off between the two DEG CPCs was calculated using the difference in the half maximum times of the PSM and the TSI CPC together with the growth

rates. The statement was made more clear in the revised manuscript.

After the sentence, P. 8, I. 11-14: A similar method for establishing GRs from CPCs with different cut-off sizes is described in Riccobono et al., 2012 and Lehtipalo et al., 2014. we added:

The cut size of the DEG CPC 2 could not be determined via calibration measurements using the CLOUD chamber. The cut-off diameter was estimated to be 0.3 nm smaller than the DEG CPC 1, based on the growth rates calculated using different ultrafine CPCs and their respective cut-off diameters (PSM  $d_{50}$ =1.1nm, TSI 3776  $d_{50}$ =2.9nm).

**Comment 20**. P. 12803, I. 25: "The GRs from the PSMS were determined in the same manner." What is the impact of the lowered ambient temperature on the cut size of the PSMs that were used to determine growth rates? What is the effect of temperature-dependent PSM cut sizes on measured growth rates? I suspect there is an impact since the main thrust of this paper is that the DEG CPC cut size is temperature dependent. If the PSM cut size is not temperature dependent, please provide a justification.

**Reply:** as already stated, the PSM cut sizes were adjusted according to the same homogeneous background for each temperature setting, assuming that this gives a reasonably similar cut size. We clarified that in the revised manuscript as shown in the reply to Comment 12 of the referee.

**Comment 21**. P. 12804, I. 1 - 5: It is unclear why the growth rates should be normalized to the same sulfuric acid concentration. Please provide a further explanation/reference/justification for this procedure.

**Reply:** the nucleation experiments in CLOUD are performed at various sulfuric acid concentrations and temperatures for the results presented in the manuscript. In order to determine whether the GRs are temperature dependent, the various sulfuric acid concentrations from the experiments used for the analysis were normalized to a fixed SA concentration  $(10^7/cm^3)$ . This was done by multiplying the calculated GR which are a function of the SA concentration with  $10^7$  and dividing that by the measured SA conc. This is a very simple method, assuming a linear relationship between the GR and the SA concentration. We clarified this point in the revised manuscript as follows:

P. 8, I. 24- P. 9, I.5: The nucleation experiments in CLOUD which were used for the GR calculations shown here, were done at different temperatures and sulfuric acid concentrations. To determine a possible temperature trend for the different instruments, the growth rates shown here were normalized to the same sulfuric acid concentration (10<sup>7</sup> cm<sup>-3</sup>) This was done by multiplying the calculated growth rates, which are a function of the sulfuric acid concentration with 10<sup>7</sup>. This factor was then dividing the by the measured sulfuric acid

concentration. That procedure was applied for each growth rate/SA concentration pair individually (see Equation 1). This is a very simplified method based on the assumption that there is a linear relationship between the growth rates and the sulfuric acid concentration. From the measurements with a high-resolution mass spectrometer, it is known that at temperatures below 278 K, the major species contributing to the nucleation and growth in the CLOUD chamber, is sulfuric acid (Schobesberger et al., 2013).

**Comment 22**. P. 12804, I. 11: Which of the two DEG CPCs is associated with the calibration curves in Figure 3 – DEG CPC 1 or DEG CPC 2? Also, please present the calibration data for both DEG CPCs since both DEG CPCs operate at different cut sizes. Any growth rate calculation based on the rise times and cut sizes between DEG CPC 1 and DEG CPC 2 requires that the temperature dependent cut sizes of both DEG CPCs be characterized.

**Reply:** DEG CPC1 is associated with the calibration curves in Figure 3. We added a description in the Figure caption, as follows:

P. 22: The blue, green and red curves show fits to the results from calibration measurements for the DEG-CPC 1, done at CLOUD in fall 2011 and the symbols show the measured data points, ...

For the low temperature measurements, we do not have a set of calibration experiments for DEG CPC 2. Therefore we estimated the difference in the cut size of the two DEG CPCs based on the back calculation from the difference in the half maxima of the PSM and the TSI 3776 CPC. This fact is explained in the revised manuscript

in section: Method for determining growth rates from the DEG CPCs as follows:

P. 7, I.22-26: To determine whether the shift in the cut-off curves influences the scientific outcome of the measurements with the laminar flow DEG CPCs significantly, the growth rates have been determined and compared to other instruments (NAIS, Mirme et al, 2013) and PSM). The growth rates from the PSM were determined by using two PSMs at different cut sizes together with the other ultrafine CPCs.

**Comment 23.** P. 12804, I. 15 - 16: What is the justification for using this particular fitting equation as opposed to others (provide a reference for this particular fit)? What is the physical meaning of parameter c? Also, as mentioned earlier, each data point in Figure 3 also has a spread in mobility diameter due to the finite width of the nano-DMA transfer function. Please include the impact of this uncertainty on the resulting parameter fit.

**Reply:** Reference to the fit is given in the revised manuscript (Stolzenburg and McMurry, 1991) Parameter c is the rate of change (/nm). The error

estimation on the x-axis is based on the measurements by Jiang et al, 2011 and are 0.185 at 1.47 nm, 0.158 at 1.78 nm and 0.14 at 3 nm for room temperature. We can assume that the error due to the transfer function (which is mainly due to diffusion losses) is even smaller for the low temperature measurements. These were not included in the fits.

**Comment 24.** P. 12805, I. 1: Low particle concentrations indicate low particle counts, which is a measure of absolute uncertainty in a single measurement (Poisson noise), not relative uncertainty (which would be uncertainty associated with repeated measurements). In Figure 3, do the counting efficiency uncertainties (vertical) refer to uncertainties associated with Poisson noise (counts), or to uncertainties associated with an average of several measurements?

**Reply:** the uncertainties are associated with an average of several measurements. For each of the calibration measurements, a series of data points were taken at the same size, as we were repeatedly stepping through the voltages with the DMA. So the resulting vertical errors are relative errors.

**Comment 25.** P. 12805, I. 3 - 5: If there were potential evaporation of the sampled aerosol, this effect of evaporation should also be present in the reference PSM counter. What impact would this have on the interpretation of the detection efficiency data?

Also, if particle evaporation is used as an explanation for the change in cut-off size, then the reduced sampling temperature is not so much having an effect on the super- saturation profile in the condenser as it is on reducing particle size before the particle reaches the zone of super-saturation/activation. In this case, the effect of temperature is not to increase the cut-off size (which is a property of the CPC), but to change the particle size before it is activated in the CPC. In other words, the CPC cut-off size would not change as a function of temperature. Please amend the text to reflect this point.

Again, as mentioned earlier, it would be very instructive (if not necessary) to see what impact (if any) a reduced sampling temperature would have on the instrument super- saturation from a model/theoretical prediction to help understand these results.

**Reply:** The background of the PSM that originates from homogeneous nucleation was kept constant for different chamber temperatures, by adjusting the temperature settings of the PSM when the chamber temperature was changed to the same background count rate. This suggests that effective supersaturation inside the reference instrument was stable. We can further assume that if the effective supersaturation stays constant for different experimental temperatures, also the counting efficiency curves stay constant.

This is of course not the optimal solution, as is using the PSM as a reference for calibration measurements, nevertheless it was the only option for those experiments. That issue needs some deeper investigation, which is beyond the scope of this paper.

We agree with the referee. If the particles evaporate before they are activated in the CPC they would become smaller, therefore they will be detected at a different size than under normal conditions, resulting in a cut-off shift. This occurs after the particles were size selected in the DMA, as until then the setup is in a cold environment. The CPC cut-off size would still be changing with temperature, as if there is evaporation, it would depend on the difference in temperature the particles experience. We addressed these points in the revised manuscript as follows:

In section: upper tropospheric temperature calibration results, P. 10, I. 17-P.11, I. 1: Two effects could cause the shift in the counting efficiency curves. The first one, is a possible change in the profile of supersaturation in the condenser of the CPC, due to a different temperature of the sample flow. In that case, the shift would be expected to be a function of temperature. Modelling results, using COMSOL 5.0 multiphysics model show that assuming a constant flow of 0.3 lpm from the inlet of the CPC until the beginning of the capillary, the final temperature of the flow is 326 K for an inlet temperature of 208.15 K and the same for an inlet temperature of 298.15 K. The dimensions that were used for the modelling are: 2.2 cm inlet at ambient temperature, 17.3 cm inside the CPC cabin, assuming at temperature of 303.15 K and 2.7 cm inside the saturated region, assuming a temperature of 328.15 K. These results suggest that the ambient temperature does not influence the profile of the saturation ratio in the condenser of the CPC. The second effect would be that the shift is caused by evaporation of particles before being activated in the CPC. This means that, after the particles are size-selected in the DMA, which is still in the cold environment, they shrink. That leads to a shift in the counting efficiency as the particles changed in size after they were size selected.

**Comment 26.** P. 12805, I. 19 - 20: Again, please provide further explanation/justification of normalizing the growth rates to a fixed sulfuric acid concentration.

**Reply:** The text in the revised paper was changed as follows, P. 8, I. 24- P. 9, I.5: The nucleation experiments in CLOUD which were used for the GR calculations shown here, were done at different temperatures and sulfuric acid concentrations. To determine a possible temperature trend for the different instruments, the growth rates shown here were normalized to the same sulfuric acid concentration (10<sup>7</sup> cm<sup>-3</sup>) This was done by multiplying the calculated growth rates, which are a function of the sulfuric acid concentration

with 10<sup>7</sup>. This factor was then dividing the by the measured sulfuric acid concentration. That procedure was applied for each growth rate/SA concentration pair individually (see Equation 1). This is a very simplified method based on the assumption that there is a linear relationship between the growth rates and the sulfuric acid concentration. From the measurements with a high-resolution mass spectrometer, it is known that at temperatures below 278 K, the major species contributing to the nucleation and growth in the CLOUD chamber, is sulfuric acid (Schobesberger et al., 2013).

**Comment 27.** P. 12806, I. 21 - 23: This statement is a bit confusing: "This equation allows the evaporation of particles if the vapour pressure of condensing vapour is low, i.e. evaporation dominates over condensation." Consider re-wording the sentence to read "This equation accounts (or allows for) for the evaporation of particles." Also, the fact that the vapor pressure of the condensing species is low has no impact on whether evaporation does/or does not occur, but only impacts the relative contribution of evaporation to condensation.

**Reply:** We thank the referee for the suggestion. We changed the sentences as follows: P. 12, I. 12-14: This equation accounts for the evaporation of particles. The relative contribution of evaporation versus condensation, is determined by the vapour pressure of the condensing species.