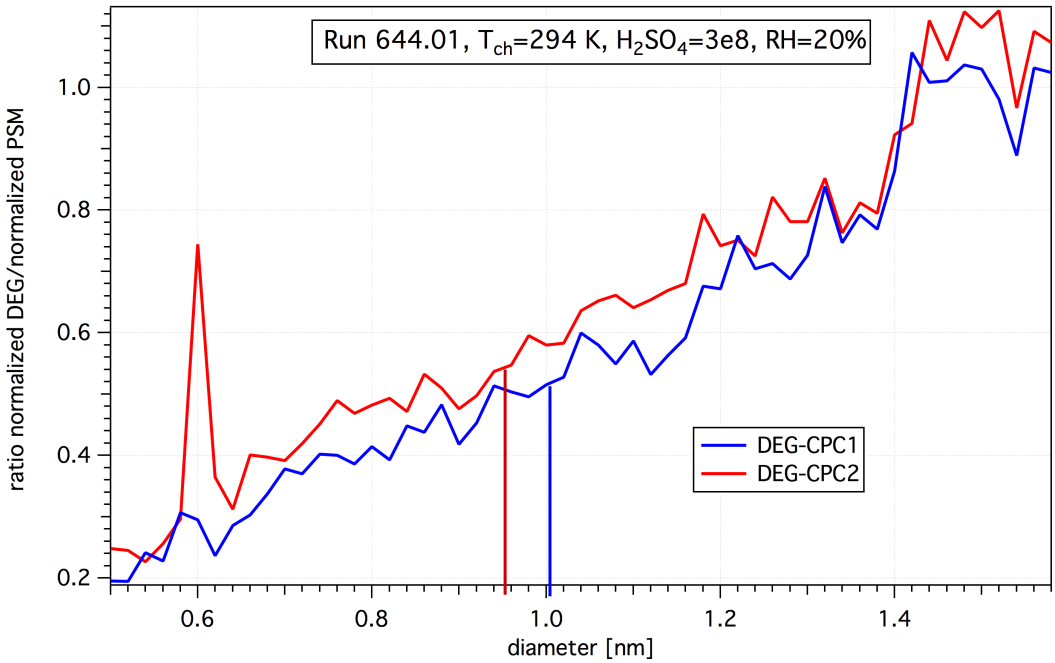
***Reply to interactive comment from Referee #1 on*: 'Technical Note: Using DEG CPCs at upper tropospheric temperatures' by D. Wimmer et al.**

We thank the referee for her/his review and comments on the draft that improved the manuscript considerably. We will address the comments point-by-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, l. 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, l.,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.

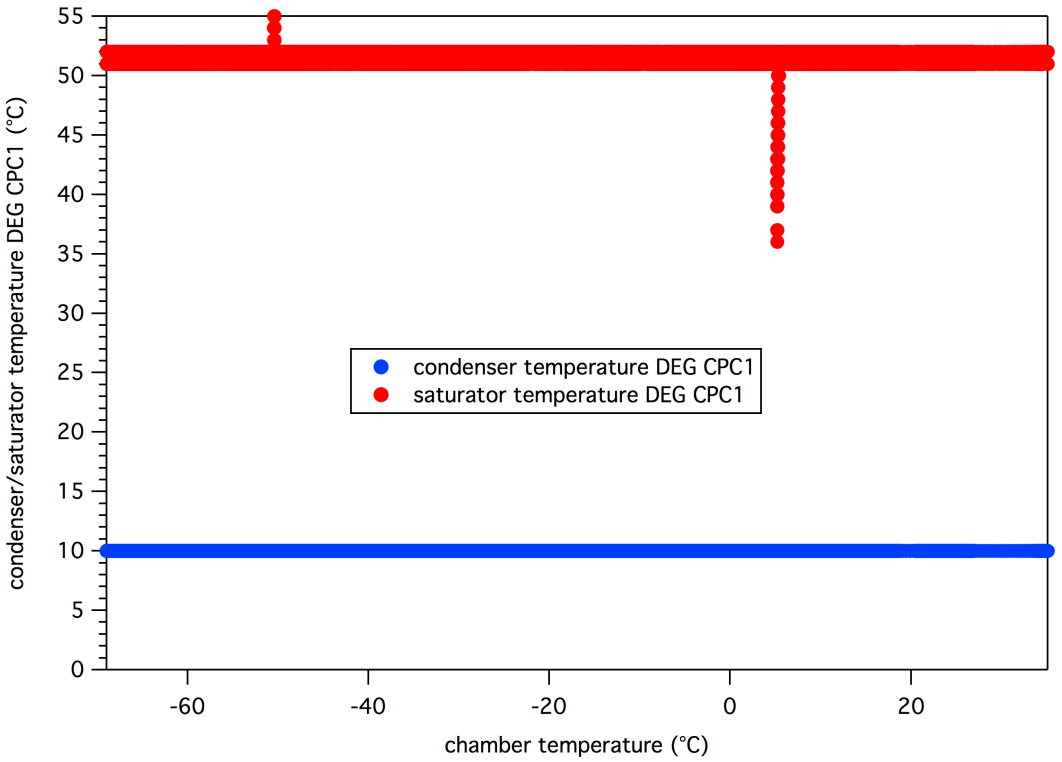
Reply: The calibration measurements using the CLOUD chamber as aerosol generation unit were done by doing several consecutive runs at the same settings. For the results at -50.5°C and -26.9°C one run was used, whereas for the results at -65.9 two sets of measurements were used, which results in different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

Reply: 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