

Interactive comment on "Technical Note: Using DEG CPCs at upper tropospheric temperatures" *by* D. Wimmer et al.

Anonymous Referee #2

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Summary: This manuscript presents results from a study that seeks to understand (1) the effect of reduced sampling temperature (representative of the upper troposphere) on the performance of sub 3 nm diethylene glycol based condensation particle counters (CPCs) and (2) what impact (if any) this temperature dependent instrument performance has on the determination of aerosol growth rates. This study is novel in that it attempts to use the CERN CLOUD chamber as an aerosol source for instrument characterization, and that it presents the first characterization of sub 3 nm CPCs at sample temperatures representative of the upper troposphere. However, the manuscript as it stands requires both substantial revisions and some further studies before it can be recommended for publication.

Comments: 1. P. 12799, I. 1 – 2: The definition of cut-off diameter should be clarified C4476

to "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.

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.

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

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

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

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).

7. P. 12799, l. 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.

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.

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.

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.

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.

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 temperature, then its use as a true reference counter at different 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".

13. P. 12802, I. 16 – 21: Is the UV turned off after the particles "grow for a couple of

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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.

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).

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".

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)?

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.

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.

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.

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.

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.

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.

23. P. 12804, l. 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.

24. P. 12805, I. 1: Low particle concentrations indicate low particle counts, which is a

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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?

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 supersaturation 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 supersaturation from a model/theoretical prediction to help understand these results.

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

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.

References

lida, K., et al. (2009). "Effect of working fluid on sub-2 nm particle detection with a laminar flow ultrafine condensation particle counter." Aerosol Science and Technology 43(1): 81-96.

Jiang, J., et al. (2011). "Transfer functions and penetrations of five differential mobility analyzers for sub-2 nm particle classification." Aerosol Science and Technology 45(4): 480-492.

Kangasluoma, J., et al. (2014). "Sub-3 nm particle size and composition dependent response of a nano-CPC battery." Atmos. Meas. Tech. 7(3): 689-700.

Kulmala, M., et al. (2007). "The condensation particle counter battery (CPCB): A new tool to investigate the activation properties of nanoparticles." Journal of Aerosol Science 38(3): 289-304.

Sipilä, M., et al. (2009). "Laboratory Verification of PH-CPC's Ability to Monitor Atmospheric Sub-3 nm Clusters." Aerosol Science and Technology 43(2): 126-135.

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 12797, 2014.

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