Author Comment to Reviewer 1

Manuscript title: "*Evaluation of DMSO as Working Fluid in Condensation Particle Counters*"
Manuscript number: egusphere-2026-127

Below, we respond to each comment point by point. Reviewer comments are presented in *italic*, followed by our responses in green. Changes in the revised manuscript are highlighted in red, with line numbers referring to the annotated manuscript.

Reviewer Comments

The manuscript by Kirchhoff et al. describes a comprehensive study on the performance of a Grimm Sky CPC with DMSO as an alternative working fluid to butanol. DMSO has several advantages over butanol, among others, it is non-toxic, not flammable and is consumed significantly less during CPC operation. It is therefore a very promising candidate for airborne measurements. In line with this potential application, the authors investigated the use of a CPC with DMSO under various pressure levels from atmospheric pressure down to 250 hPa. They found the performance to be comparable to a butanol CPC, but also noticed that the droplet growth is clearly different with DMSO compared to butanol, resulting in a larger number of (optically) smaller droplets. Numerical simulations were conducted to explain the distribution of super-saturation levels in the saturator in order to better understand the differences.

The manuscript is well written and presents a lot of novel information, which is certainly of interest to the readers of AMT. The manuscript merits publication, but I suggest a few minor revisions before it can finally be accepted.

We thank the reviewer for the positive and encouraging assessment of our manuscript and for recognizing the novelty and relevance of our work. We appreciate the reviewer's constructive feedback and have carefully addressed all comments and suggestions in the revised version of the manuscript. All changes are detailed in the responses below.

1. Introduction, general: What I miss is a brief introduction into what type of aircraft measurements the CPC is intended to be used for, i.e. at what altitudes and corresponding pressure levels.

We agree that providing more context on the intended application improves the clarity of the manuscript. We have therefore added a brief description of typical aircraft-born measurement conditions, including relevant altitude ranges and corresponding pressure levels, to the Introduction. This addition helps to better frame the investigated pressure range in the context of airborne applications.

L.52 ff: In the context of the IAGOS infrastructure (In-Service Aircraft for a Global Observing System, Petzold, A. et al. (2015)), instruments are operated onboard passenger aircraft over extended periods of several months and are exposed to a wide range of ambient conditions, including cruise altitudes of approximately 8–12 km, corresponding to pressure levels of about 200–300 hPa, as well as near-surface conditions during ascent and descent.

2. Page 2, second paragraph: I suggest to add the information that CPCs are among the very few aerosol measurement techniques that are metrologically traceable (ISO 15900).

We have added a statement highlighting that CPCs are among the very few aerosol measurement techniques that are metrologically traceable in accordance with ISO 27891:201, since the ISO 15900 suggested by the reviewer is the standard for Differential Electrical Mobility Analysis. This addition clarifies the reliability and accuracy of CPC measurements. The revised text can be found in the Introduction.

L. 29 f: CPCs can detect particles as small as a few nanometers in diameter, and they are among the very few aerosol measurement techniques that are metrologically traceable in accordance with ISO 27891:201, which ensures high confidence in measurement accuracy.

3. Page 2, line 47: Why is the higher liquid consumption of a water CPC a particular limitation for aircraft measurements? From my personal experience, water CPCs can be operated for days if not weeks before the water reservoir needs to be refilled. One main disadvantage of water CPCs is, however, not mentioned and that is its material dependence, especially of the cut-off diameter.

We thank the reviewer for this valuable comment and for sharing their experience. We agree that water-based CPCs can typically be operated for several days to weeks without refilling under standard conditions. We have clarified this point in the revised manuscript and softened our original statement. However, in the context of long-term autonomous operation onboard passenger aircraft (e.g., within the IAGOS infrastructure), instruments are deployed for periods of several months without maintenance. Under these conditions, high working fluid consumption can become a practical limitation, particularly for instruments without active fluid recycling. While newer water-based CPC designs may include such recycling mechanisms, the CPC configuration used in this study does not.

We further appreciate the reviewer's important remark regarding the material dependence of water-based CPCs. We agree that the activation efficiency and cutoff diameter depends on the properties of the particles themselves, which is more pronounced for water-based systems. This aspect has now been included in the revised manuscript.

Reviewer 2 raised similar concerns, which have also been addressed. The corresponding revisions have been incorporated into the Introduction.

L. 57 ff: One promising alternative working fluid is water, as employed in modern-generation CPCs (Hering et al., 2014). Using water as a working fluid avoids the health and safety concerns associated with alcohols. However, certain practical aspects may limit its suitability for long-term autonomous aircraft operation. While modern water-based CPCs can operate with very low working fluid consumption under typical ambient conditions, consumption may still increase depending on instrument design and operating conditions, which can become relevant when maintenance or refilling is not feasible over extended deployment periods. In addition, contamination effects during prolonged inactivity require careful mitigation strategies. Furthermore, water-based CPCs can exhibit a stronger dependence of activation behavior and cutoff diameter on particle material, which may introduce additional uncertainties in particle detection under varying conditions (Mei et al., 2021; Weber et al., 2022).

4. Page 9, section 3.2: The use of symbols is quite confusing. According to Figure 3, c0 and c1 are voltages. In the text of section 3.2, all of a sudden Th appears as a symbol for a threshold voltage. It took my quite some time to realize that c0 and c1 are actually concentrations with c1 being the concentration of particles/droplets deemed valid, based on the fact that the scattered light peak triggered a voltage signal, exceeding a defined threshold Th(c1). c0 is apparently the concentration of all droplets that cause a signal above noise level. Figure 3 and section 3.2 should be rewritten to make this clear and not leave the understanding up to the interpretation of the reader. I also wonder if Th is an appropriate symbol. Shouldn't this rather be something like Uth to make it clearer that it is a voltage threshold?

We agree and have revised Section 3.2 to clarify the meaning of c_0 and c_1, which are internal parameters of the CPC manufacturer. For a better understanding we newly introduce the voltage thresholds $U_{\mathrm{th}}(c_0)$ and $U_{\mathrm{th}}(c_1)$. In the revised text, $U_{\mathrm{th}}(c_0)$ and $U_{\mathrm{th}}(c_1)$ explicitly indicate the internal voltage thresholds corresponding to each particle number concentration. We also clarified that c_1 represents fully grown droplets exceeding the higher threshold, while c_0 includes all particles above the lower threshold (noise) but below $U_{\mathrm{th}}(c_1)$. Figure 3 has also been updated to improve clarity and ensure that the definitions of the symbols and thresholds are immediately understandable. These changes aim to eliminate any ambiguity in the representation of c_0, c_1, and the associated voltage thresholds.

L. 230 ff: The Sky-CPC 5411 provides an internal diagnostic of droplet growth. Aerosol particles enter the instrument, undergo condensational growth to droplets in the condenser, and are subsequently detected optically. This growth process is evaluated by the instrument using two fixed signal thresholds.

In the following, we refer to the particle number concentrations c_0 and c_1, which the instrument derives from two fixed manufacturer-defined voltage thresholds in order to distinguish different levels of droplet growth based on the scattered-light signal. These thresholds are $U_{\mathrm{th}}(c_0) = 0.5\,\mathrm{V}$ and $U_{\mathrm{th}}(c_1) = 1.2\,\mathrm{V}$, respectively.

The lower threshold $U_{\mathrm{th}}(c_0)$ represents the minimum signal required for a grown droplet to be detected above the noise level. The corresponding concentration c_0 therefore includes all aerosol particles that have grown into optically detectable droplets. The higher threshold $U_{\mathrm{th}}(c_1)$ requires a stronger scattered-light signal and is only exceeded by droplets that have grown to larger sizes. The concentration c_1 thus represents the subset of detected droplets that produce a higher optical signal. By design, the instrument reports the c_1 concentration as the particle number concentration.

The ratio c_1/c_0, hereafter referred to as the *indicative droplet growth ratio*, provides a diagnostic of the growth efficiency, i.e. the fraction of optically detected droplets that reach the higher signal regime within each 1 s measurement interval. Values of $c_1/c_0 < 1$ indicate that a fraction of grown droplets does not reach the higher detection threshold.

As illustrated in Fig. 3, the DMSO droplet diameters corresponding to the c_0 and c_1 thresholds are approximately $2.5\,\mu\mathrm{m}$ and $4.6\,\mu\mathrm{m}$, respectively, with smaller droplets not being counted. These values were derived by (Weber et al., 2023)from signal measurements of latex test particles combined with a model of scattered-light intensity for the GRIMM measuring cell.

Since these thresholds were originally defined by the manufacturer for operation with butanol, an adjustment is required when using DMSO. This is due to the lower vapor pressure of DMSO, which reduces the available vapor for condensational particle growth. Ideally, this adjustment would be implemented in the instrument hardware through a redefinition of the voltage thresholds for DMSO operation. As a practical workaround in this study, we use the c_0 count as the particle number concentration.

5. Page 9/10, section 3.3, including Figure 4: How has this been measured? With monodisperse, DMA-classified particles? If so, for which size(s)? Or was it a polydisperse aerosol? If so, how did you know the charge level to obtain the reference number concentration from the FCE?

The measurements in Figure 4 were performed using NaCl particles (generated with a nebulizer and classified with the DMA as shown in the setup in Fig. 1). A single DMA scan cycle was conducted, covering particle diameters from 140 nm down to 2.5 nm, yielding quasi-monodisperse aerosol populations at each mobility step. A multiple charge correction to was applied.

L. 270 ff: The measurement shown in Fig. 4 was performed using the experimental setup described in Sec. 2.2.1 and illustrated in Fig. 1. A single DMA scan cycle was conducted, covering particle diameters from 140 nm down to 2.5 nm, producing monodisperse aerosol populations at each step. The reference number concentration for each size was determined using the FCE, which measures particle charge to provide an accurate, traceable count.

6. Page 12, Figure 6: In the caption of the figure and the x-axis, it should read surface area concentration rather than just surface area. What test aerosol was used for these measurements? How has the surface area calculated? Assuming that this result was achieved for rather compact (NaCl or AS) particles, what would happen if particles are highly agglomerated (e.g. soot), thus with a much higher surface area per particle for the same equivalent diameter?

The x-axis, figure caption and corresponding text passages have been updated to indicate surface area concentration. The measurements were performed using NaCl particles as the test aerosol. Surface area concentration was calculated by multiplying the number concentration by the surface area of a sphere with radius determined from the DMA classification.

Measurements with highly agglomerated soot particles exhibit the same overall behavior (Fig. 1), demonstrating that the CPC response is robust for both compact and agglomerated aerosols. The small dip at optimal temperature settings observed at the very beginning of each measurement cycle occurs in almost all measurements, independent of particle type, and is therefore likely attributable to an artifact in the measurement or experimental setup rather than a physical effect. These clarifications have been incorporated.

To enhance the overall clarity of this section, we have included a more detailed discussion of the results.

L. 319 f: The surface area concentration was calculated by multiplying the number concentration by the surface area of a sphere with radius determined from the DMA classification. [...]

L. 341 ff: This observed behavior can be understood as the result of coupled particle-size-dependent activation and finite residence time in the condenser. Larger particles require lower supersaturation

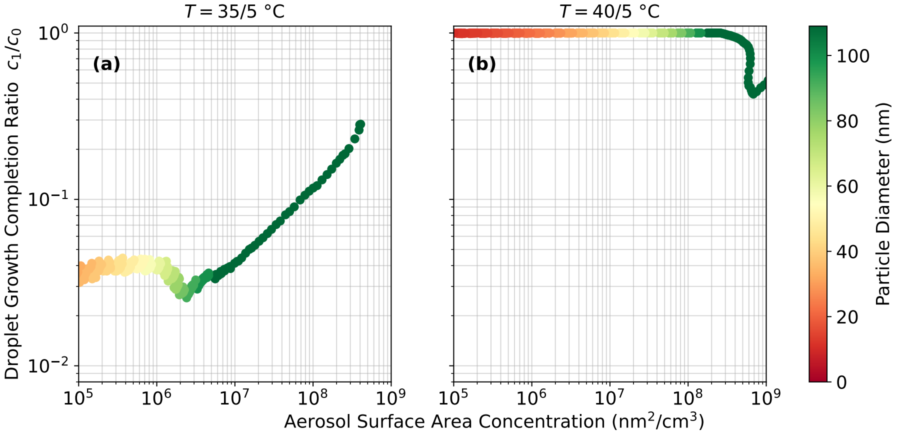

Figure 1: Indicative droplet growth ratios c_1/c_0 versus total aerosol surface area concentration for soot aerosols at 1000 hPa. (a) Non-optimal temperature setting: $T_{\text{sat}} = 35\,^{\circ}\text{C}$, $T_{\text{con}} = 5\,^{\circ}\text{C}$ (left). (b) Optimal temperature setting: $T_{\text{sat}} = 40\,^{\circ}\text{C}$, $T_{\text{con}} = 5\,^{\circ}\text{C}$.

for activation and can therefore activate earlier along the condenser, resulting in longer effective growth times compared to particles near the cutoff diameter. This size dependence is consistent with the observed variation of c_1/c_0 with particle diameter.

Furthermore, measurements with highly agglomerated soot particles exhibit the same overall behavior, demonstrating that the CPC response is robust for both compact (NaCl) and agglomerated (soot) aerosols with highly different surface properties.

The representation as a function of aerosol surface area concentration further highlights this behavior. At low surface area concentrations, the system operates in a regime where droplet growth is primarily limited by insufficient residence time, i.e. a time-limited regime. This can equivalently be interpreted as the effective condenser length being insufficient for complete droplet growth under the given operating conditions. At higher surface area concentrations, deviations from this behavior would indicate that vapor depletion may begin to contribute, suggesting a transition toward a vapor-influenced regime.

7. Page 16, Figure 9: Why does the tube length start at 110 mm? Do you only show a part of the saturator or does the section from 0-110 mm belong to the condenser? How did you derive the saturation profile at the inlet of the saturator?

Figure 9 only shows the condenser section of the CPC, starting at 110 mm from the instrument inlet. The preceding parts (saturator: 0–100 mm and insulator: 100–110 nm) are not shown, as our focus is on supersaturation within the condenser and the droplet growth at its outlet. The saturation profile at the condenser inlet was derived from the simulated temperature and water vapor distribution at the end of the saturator, including the insulating section (as described in Sec. 2.2).

L. 420 ff: Figure 9 only shows the condenser section of the CPC, starting at 110 mm from the instrument inlet. The preceding parts, including the saturator (0 – 100 mm) and the 10 mm insulating section, are not shown, as the focus is on supersaturation within the condenser and the droplet growth at its outlet.

8. Page 21, Figure 14: x- and y-axis are reversed. The D_{50} is a function of the temperature difference and not vice versa.

We agree that, strictly speaking, the D_{50} is a function of the temperature difference. However, we have chosen to present the data with particle diameter on the x-axis to improve readability (see below) and facilitate comparison with other figures, since the particle size is usually shown on the x-axis. In this way, the dependence of D_{50} on temperature difference can be more intuitively interpreted.

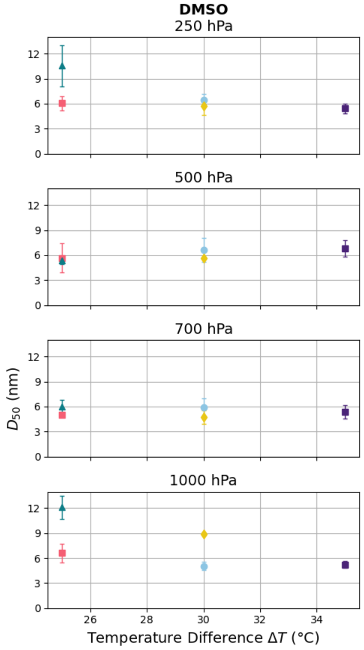

Figure 2: Same dataset as presented in Fig. 14 of the original manuscript, with axes exchanged.

Page 24, Figure 16: Earlier, you mention that the POPS measurements showed smaller droplet diameters for DMSO than for butanol and speculated that this may be due to the difference in the refractive indices. Could this also be the reason for the lower c1/c0 ratios, because the corresponding thresholds are for the voltages, which reflect the peak height of the scattered light in the optical detection of the CPC?

While differences in refractive index do influence the scattered light intensity, a higher refractive index leads to stronger light scattering for droplets of the same size. In this context, DMSO has a refractive index of approximately $n \approx 1.48$, compared to $n \approx 1.40$ for n-butanol (at $\lambda = 589$,nm, $T \approx 20°\mathrm{C}$). Therefore, if droplet sizes were identical, DMSO droplets would be expected to produce higher, not lower, signal peak heights.

Consequently, the refractive index difference alone cannot explain the observed lower c_1/c_0 ratios. Instead, we attribute this behavior primarily to the smaller droplet diameters observed for DMSO. Since the scattered light intensity strongly depends on particle size, even small reductions in droplet diameter lead to significantly lower signal amplitudes. As a result, a larger fraction of DMSO droplets does not exceed the higher detection threshold $U_{\mathrm{th}}(c_1)$, leading to reduced c_1/c_0 ratios.