Author Comment to Reviewer 2

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 authors present data and modeling of a condensation particle counter operated with DMSO as its working fluid. This is an interesting idea, and well-deserving of the further exploration presented here. The experiments are carefully conducted, and apart from a few points listed below, the results are clearly presented. Yet there are several significant points the authors should address prior to publication:

We thank the reviewer for the careful evaluation of our manuscript and for the constructive and encouraging feedback. We appreciate the recognition of the novelty of operating a condensation particle counter with DMSO as a working fluid, as well as the acknowledgment that the experiments were conducted with care and that the results are, for the most part, clearly presented. We have carefully considered all comments and suggestions and have revised the manuscript accordingly. Below, we provide a detailed, point-by-point response to the reviewer's remarks. All changes made in the manuscript are highlighted accordingly.

1. This Reviewer's primary concern is that the instrument itself, the DMSO CPC, is not described. Nor is it described in the paper Weber et al (2023) "A new working fluid...,.", which presumably the authors meant to cite. The authors do state they have used a commercial instrument with DMSO instead of butanol. Yet, as this is an archival journal, it is important to describe the pertinent physical characteristics of that instrument. A model number will not suffice even 10 years from now. The authors might include a sketch, and at a minimum give some of the critical dimensions, such as for the saturator and condenser length, the orientation of the condenser with respect to the saturator. They should give the flow rate, and state how this flow is adjusted as a function of inlet pressure. Did they use a constant volumetric flow, a constant air mass flow or something else? Is the instrument sheathed? One infers from the model results of Figure 9 that this is a laminar flow instrument operated at constant volumetric flow, but the authors should so state.

We agree that the description of the CPC instrument in the original manuscript was not sufficiently detailed for an archival publication. In the revised manuscript, we have therefore significantly expanded the instrument description and now include the relevant physical and operational characteristics of the CPC, such as flow conditions and key design aspects.

L. 130 ff: Two Sky-CPC 5411 instruments (GRIMM Aerosol Technik) were used for all measurements. The Sky-CPC is a commercially available condensation particle counter designed for aviation applications. It is based on a laminar-flow, continuous-flow CPC design consisting of a saturator and a downstream condenser. The aerosol flow is conditioned in the saturator, where it becomes saturated with working fluid vapor, and subsequently enters the condenser, where a temperature gradient induces supersaturation and particle activation followed by droplet growth.

In this study, one CPC was operated with butanol (B-CPC) as intended by the manufacturer, while the second instrument was operated with dimethyl sulfoxide (DMSO) as working fluid (DMSO-CPC). Both instruments were operated at a constant volumetric flow rate of $Q = 0.6\,\text{LPM}$, resulting in laminar flow conditions within the growth tube. As ambient pressure changes, the corresponding mass flow rate and residence time inside the condenser vary accordingly. The flow configuration and general instrument design follow the description given in Bundke et al. (2015).

2. The cited reference for Weber et al (20203) is to "Characterization of a self-sustained, water-based condensation particle counter for aircraft cruising pressure level operation", AMT 16:3505-3514). One assumes the authors meant to cite Weber et al (2023) "A new working fluid..,." Aerosol Research 1, 1-12, 2023. Please add the correct reference.

The reference was indeed incorrect. We have corrected the citation to "A new working fluid for condensation particle counters" in the revised manuscript.

Weber, P., Bischof, O. F., Fischer, B., Berg, M., Schmitt, J., Steiner, G., Keck, L., Petzold, A., and Bundke, U.: A new working fluid for condensation particle counters for use in sensitive working environments, Aerosol Research, 1, 1–12, https://doi.org/10.5194/ar-1-1-2023, 2023b

3. Given the overlap in authorship, it is very strange that the results from Weber et al (2023) is "a self-sustained, water-based… for aircraft" are not included in Table 1. That paper has results for both butanol and water-based instruments. This reviewer recommends that these results be included.

We agree that the results from "Characterization of a self-sustained, water-based condensation particle counter for aircraft cruising pressure level operation" are relevant in this context. We have therefore included the corresponding data for both butanol- and water-based instruments in Table 1 in the revised manuscript. For improved readability of the table, we excluded two columns of the originally submitted manuscript that did not provide substantial further information.

The annotated manuscript directly contains the updated version of the table, rather than a change-marked comparison, as the latter would significantly reduce readability.

4. The statements about the disadvantages of water as a working fluid are gratuitous and should be struck. First, it is simply not true that the high diffusivity of water leads to greater water consumption. Water-based condensation particle counters have been designed that consume little to no water – under typical ambient conditions these instruments operate for weeks to months without consuming any water at all. Second, organic or biological contamination can be handled, just as manufacturer's have solved the problem of water-uptake by the wick of butanol instruments that killed the performance in the original designs. (how hard would it be to add an activated carbon filter in the water fill line?). Further, not only is water readily available, nontoxic and non-flammable, note that the water vapor saturation ratio required for the activation of 5nm particles is less than one-half of that required when using butanol or DMSO (by the Kelvin relation the critical saturation ratio depends on the product of molecular weight, surface tension divided by the liquid density, which for water is less than one-half of that of butanol).

We agree that some of the statements regarding the disadvantages of water as a working fluid were overly general and not sufficiently nuanced. In particular, we acknowledge that modern water-based condensation particle counters can achieve very low water consumption and that solutions exist to mitigate contamination effects.

We have therefore revised the manuscript to soften and clarify these statements, avoiding overly broad claims. At the same time, we retain a brief discussion of practical challenges associated with water-based systems in specific operating conditions, as these considerations are relevant to the context of this study. For completeness and to improve the clarity of the study motivation, Fluorinert FC-43 is also briefly included in the discussion of alternative working fluids.

Reviewer 1 raised similar concerns, which have also been addressed. The corresponding revisions have been incorporated into the Introduction.

L. 56 ff: An alternative working fluid that has been used in some low-pressure applications is Fluorinert FC-43 (Brock et al., 2000; Richter et al., 2026). However, its use is limited to very low ambient pressures and it is not considered environmentally sustainable due to its high global warming potential and persistence in the atmosphere (Hong et al., 2013). For these reasons, it is not suitable for routine atmospheric measurements in aircraft-based observing systems. [...]

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 (Mei et al., 2021), 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).

5. Section 3: This Reviewer appreciates the thoroughness with which the data have been handled. However, this section would be clearer if it were written more concisely – It isn't until the last sentence of Section 3.2 that the reader knows what is meant by "adjustment to Co threshold" in Figure 2 that is referenced ahead of Section 3.1.

Reviewer 1 raised the same point, and we have revised the entire paragraph to improve clarity and conciseness.

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 $1\,\mathrm{s}$ 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.

6. Section 3.3—what particle diameter was used for these tests? Or, if a polydisperse aerosol, what was its approximate mean diameter?

Reviewer 1 raised the same point: 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.

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.

8. Section 4.2. It would be good to point out the fundamentals of what happens to the saturation ratio profiles at reduced pressure. In a cylindrical tube, for fixed temperatures and working fluid, the heat and mass transport vary with Dz/Q, where D is the diffusivity, Q the volumetric flow and z the axial coordinate. Both thermal and mass diffusivity are inversely proportional to the pressure. Were these profiles modeled at fixed air mass flows, they would look nearly the same. Reducing the pressure at fixed volumetric flow increases Dz/Q making the saturation peak further towards the inlet of the condenser, essentially increasing its effective length.

We agree that the fundamental dependence of the saturation profile on pressure was not sufficiently explained in the original manuscript. In the revised version, we have added a corresponding explanation, highlighting that under constant volumetric flow conditions, the increase in diffusivity with decreasing pressure enhances transport processes and shifts the supersaturation maximum toward the inlet of the condenser, effectively increasing the usable growth region.

L. 431 ff: The CPCs were operated at constant volumetric flow. As diffusivity increases with decreasing pressure, heat and mass transport within the condenser become more efficient. As a result, the supersaturation maximum shifts toward the inlet of the condenser, effectively increasing the usable length of the condenser and the residence time available for particle activation and droplet growth under reduced-pressure conditions.

9. Figure 7 shows a concentration effect, ie less droplet growth at high particle number concentrations, and this is attributed to vapor depletion. This may be correct, but why have they ruled out condensational heating as a contributor to this effect?

We agree that, in addition to vapor depletion, condensational heating can in principle contribute to reduced droplet growth at elevated particle number concentrations, as latent heat release during condensation may locally reduce supersaturation.

In the revised manuscript, we have added a brief discussion of this effect. However, a quantitative separation of these effects is beyond the scope of the present study.

L. 361 ff: This change in behavior indicates that, above a total aerosol surface area concentration of approximately $10^8\,\mathrm{nm}^2/\mathrm{cm}^3$, droplet growth becomes limited by the availability of condensable vapor, i.e. a vapor-limited regime is reached. In addition, condensational heating due to latent heat release may further reduce supersaturation at elevated particle number concentrations. However, a quantitative separation of these effects is beyond the scope of the present study.

Overall, these observations support our hypothesis that, under the conditions of the standard measurements, the observed trends in c_1/c_0 are primarily governed by a time-limited regime, in which particles do not spend sufficient time in the condenser to reach their full droplet size.

10. As a practical matter, as the condenser is generally lower than the freezing point of DMSO, what is done to prevent build up of ice on the condenser walls? Might this not be a problem for long-term operation?

In our experiments, we did not observe any indication of performance degradation that would suggest significant buildup of condensed or solidified DMSO on the condenser walls over the investigated timescales of up to a year now. Nevertheless, we itend to investigate this matter in long-run studies. Theoretically, the CPC is measuring ambient air, where water vapor is inherently present, or it can be operated using a DMSO–H_2O mixture as the working fluid. In both cases, water is present in the system. As described in Sec. 4.7, the presence of water depresses the freezing point of DMSO. Therefore, the system does not operate in a regime where DMSO freezing occurs at the condenser, and ice buildup on the condenser walls is not expected to be an issue, even during long-term operation.

L. 564 ff: Furthermore, as the CPC measures ambient air, water is always present in the system.

This lowers the effective freezing point of DMSO, such that operation at condenser temperatures below 18 °C should not result in freezing or ice buildup on the condenser walls, even during extended operation.