<u>Referee 1 comments</u>

We appreciate the thorough review and constructive feedback provided by referee 1 on our manuscript. We acknowledge the importance of addressing the issues raised and are committed to making significant improvements to enhance the clarity, readability, and scientific rigor of our work.

We acknowledge the shortcomings highlighted regarding the presentation quality of the manuscript, particularly regarding the size and readability of figures, numbering of equations, and completeness of the table of symbols. We will address these issues by improving the visual presentation of figures, ensuring the correct numbering of equations, and enhancing the completeness of the table of symbols to provide a more coherent and user-friendly reading experience.

We are grateful for the valuable feedback provided by referee 1, and we are committed to addressing the identified issues through major revisions to improve the quality and scientific integrity of our manuscript. We believe that with these proposed improvements, our work will contribute valuable insights to the field of SADS development for SVOC measurement and merit consideration for publication in a scientific journal.

The following are specific answers to the specific comments of referee 1.

RC1 : "The figures and their labeling are far too small and difficult to read, and different test results will not be clearly visible even in a black and white copy."

- Figure 1 has been split into two figures, and both parts have been enlarged for better readability.
- Figure 2, "Schematic drawing of the Bench for Organic Aerosols (BOA) generation device," has been enlarged.
- Figure 3, "Photographs of the SADS prototype, consisting of two main components - the acceleration nozzle (A’) and the collection nozzle (B’)," is considered readable as it is. No change has been made.
- Figure 4, "Schematic representation of the positions of the samplers on the crown support," is considered readable as it is. No change has been made.
- Figure 5, "Mass balance in the SADS prototypes in function of the airtightness level. Error bars represent the standard deviation calculated on five replicates for each condition," has been enlarged, and fill patterns have been added to make the figure readable in grayscale format.
- The font size of the characters in Figure 6, "Evolution of CFD mass transmission of the SADS prototypes, accounting for variation in nozzle diameters," has been enlarged for better readability.
- The font size of the characters in Figure 7, "Comparison of theoretical CFD transmission efficiency ηp (SADS R) with experimental results for SADS 1, SADS 2, and SADS R," has been enlarged for better readability.
- Figure 8, "Distribution of the fate of inertial particles with a reference diameter of 4.5 µm in multiple repetitions, for SADS 1, SADS 2, and SADS R at High Airtightness Level," has been enlarged, and fill patterns have been added to make the figure readable in grayscale format.
- Figure 9 has not been changed.

- Figure 10, "Mass distribution in SADS R exposed to four different particle size distributions," has been reviewed for better readability.
- Figure 11, "Numerical simulation results of misalignment effect compared with experimental tests," has been split into two parts called A and B, and each part has been enlarged for better readability.

RC1 : "some equations were not recorded in their number order"

- All equations were numbered.

RC1: "the table of symbols is very, very incomplete"

- The table of symbols was updated to integrate all symbols and abreviations.

RC1: "Since in the present case a modified CMAG generator was used, it would be desirable to have references to publications that demonstrate the function and the suitability of this aerosol generator, or to include a more detailed description and graphical representation in this regard in the manuscript."

- Even if our method was not inspired by (Steiner et al., 2017), the methodology is similar. As the detailed protocol is not the subject of this paper, but it is a key parameter, we wonder if a short communication paper would be profitable to present the methodology and the robustness of this method? At least, the reference to Steiner et al. was added in the main text.

Referee 2 comments

Thank you, referee 2, for your thorough review and insightful comments on our manuscript. We appreciate the time and effort you have dedicated to providing valuable feedback. Your comments highlight important areas for improvement, and we are committed to addressing each of them in the revised version of the manuscript.

General comments:

RC2: "*The overall principle of the SADS is not sufficiently described. I assume that it is based on the assumption that the relevant particles are sufficiently large to only be collected by the major flow. As long as this is the case, the minor flow would only contain the vapor phase, but no particles. The adsorbent in the major flow would contain both, the vapor, present in the aerosol as well as the vapor, evaporated from the upstream filter. The total particulate material can thus be determined from the mass collected on the filter in the major flow and the difference of the vapors on the two adsorbents. Is this understanding correct?*"

- Yes this is correct. To improve the description in the paper, we now propose to describe the SADS principle at the beginning of section 2. Section 2 has been globally rewritten to also answer other points raised during the review (see new version of section 2). Also Figure 1 has been split in two and the description of the SADS parts has been extend in the legend.

RC2: "*Information on the d50 of the virtual impactor is missing. From the experimental results, it appears to be somewhere between 0.15 µm and 2 µm. Please provide further information on this.*"

- the section 2 was completed to document these aspects (see rewritten version of section 2).

RC2: "*How realistic is an assumption that a (semivolatile) workplace aerosol only contains particles >>d50? Only with such an assumptions, the SADS principle (in my understanding, see above) could generally work.*"

- indeed, we now document this aspect in the revised form of section 2 (see rewritten version of section 2)

RC2: "*What type of adsorbents are used as samplers for the vapor phase?*"

We prefer not to lose the reader by documenting this aspect in this article. This article focuses on the effect of the mechanical design of the SADS on its internal separation performance: the question of the choice of adsorbents placed downstream of the outlets is not dealt with, because this question is disjoined from the raw separation performance of the SADS. The selection of adsorbents is addressed, for example, in standards such as EN ISO 22065, EN ISO 23861, and more broadly in EN 482, which delineate the performance tests necessary to establish a methodology for assessing worker exposure to a specific chemical agent.

RC2: "*What is the pressure drop across the acceleration nozzle and does it affect the partitioning between particulate and vapor phase?*"

- In described operating conditions, the pressure drop is 1400 Pa on the major flow side and 3700 Pa on the minor flow side. As written in the paper, the SADS are operated in constant-flow mode during experiments. In these conditions, measurements show less than respectively 7.5% and 4.7% variations of these pressure drops between prototypes, when flow rates are maintained constant as specified (see Belut et al 2022). These variations of pressure drop between prototypes are only provoked by variations of internal geometry between prototypes, that modify head losses. They are not provoked by flow rates changes.
  Thus, in presently used constant flow mode, geometry variations lead to modifications of flow patterns in the device, with subsequent modifications of pressure drops and particles partitioning. In this respect, the way that pressure drops affect particles partitioning is actually addressed in the paper.
  If your question is rather: does a change in flow rates, inducing a change in pressure drop, affect particle distribution, the answer is of course yes, but this question is not addressed in this article, which focuses on the effects of the mechanical design of the SADS on its performance, under the flow conditions for which it was designed (constant flow rates). Note that for such an incompressible flow, the pressure drop should not be considered as an univocal determinant of particle partitioning: the fate of particles in the device is determined by the flow pattern (velocity field and turbulence patterns) and geometry, not by the pressure drop per se. The flow in the device can of course be pressure driven, but it is the resulting flow pattern that determines the fate of the particles (multiple flow patterns can exist for a given pressure drop).
  To clarify this point, we propose first to insist on the fact that SADS are studied for a

constant flow rate, in section 2: “It is the details of the device's geometric dimensions and the choice of minor and major flow rates that determine the device's theoretical separation performance (Kim, Loo, Marple). In the present article, these choices are assumed to be theoretically optimal, and we study only the effects of certain design and manufacturing details on the device's ability to actually achieve its theoretical performance. Hence, minor and major flows are set constant at their theoretical optimum as specified.”
And in the conclusion: “The study was carried out for constant air flows set in accordance with SADS specifications.”
And regarding pressure drop and its general impact of the device performance, we propose to add a comment in the conclusion, linked also to your next remark (see answer to next comment).

RC2: “*It was found that the axial misalignment of the nozzles in comparison to their sizes has a major influence. Why are no larger nozzle diameters, along with a higher flow rate used?*”

- Indeed : with this question, you raise some interesting and fundamental questions about the theoretical design of the SADS. These questions go far beyond the scope of this article, which is simply to verify that the SADS design achieves its theoretical performance and to identify the causes of its poor performance. Your insightful comment highlights that the tests conducted have brought to light significant design challenges with the SADS. It appears evident that a thorough reevaluation of the separation chamber design and flow parameters is necessary, potentially involving adjustments to both nozzle diameters and flow rates, as you have suggested. Note however that the possibility of increasing the flow rates are largely limited by the performances of the personnal sampling pump, taking into account the supplementary pressure drop provoked by the collection media (adsorbent etc.). Since this issue is beyond the scope of the article, we suggest to address it in the conclusion. We therefore propose to add the following text to the conclusion:
  “*Overall, these results clearly show that it is mechanically difficult to design a SADS that meets the theoretical specifications. In fact, the alignment tolerances require precise machining, which may be an obstacle to the development of this device. It should be added that the head losses of the device at its nominal flow rate are 1400 Pa on the major flow side and 3700 Pa on the minor flow side (Belut 2022). These head losses are at the limit of the performance of individual sampling pumps, especially when considering the additional head losses caused by the collection media downstream of the SADS outlets. This raises the question of whether the device should be completely redesigned, with larger nozzle diameters that are easier to align mechanically and generate less pressure drop.*"

Specific comments:

RC2: “*Line 66: The term inertial and non-inertial particles are a bit misleading, because there are no “non-inertial” particles. I would suggest to use the terms “particles with high/low inertia”*”.

- Thank you for your comment. We have made the suggested changes to replace the terms "inertial" and "non-inertial" particles with "particles with high inertia" and "particles with low inertia" throughout the manuscript.

RC2: "*Line 75: The distinction between the two different (2009/2010) versions is a bit confusing here. It is not clear, which version has been used in this study.*"

- Indeed, this point is now specified in the introduction, and reiterated in the conclusion. It is thus now written at the end of introduction "We shall base our study on the 2009 version of the SADS, because of the smaller cut-off diameter and also considering that the issues related to the overall design of the SADS are common to both versions. To reach our objectives, five 2009 SADS prototypes were constructed..."

RC2: "*Line 76: Sequence of the figures? You refer to figure 3 before you refer to figure 2.*"

- Thank you for this comment. The citation was not appropriate and was deleted.

RC2: "*Line 119: The article not only proposes to do this, but you have done it and describe/explain it.*"

- Thank you for this comment. The paragraph was modified in response to the comment: the word 'proposes' was removed and the verbs were changed to the active voice.

RC2: "Line 154: Why do you expect ηd to be zero and not ηd + ηv? Unless all (!) particles are much larger than the d50, it should be the sum of both."

- OK You are absolutely right. The text was modified accordingly: "*In ideal working conditions of the SADS as a gas-particle separator, we expect ηd and ηv to be zero while ηp=1.*"

RC2: "*Line 183: Please provide a proper reference to the original paper by Sinclair and La Mer from 1949."*

- The citation of the original paper by Sinclair and La Mer 1949 was added properly.

RC2: "*Line 183/184: The description of the principle is too short. You should at least mention that DEHS is evaporated and then condenses onto the NaCl seed particles.*"

- Thank you for the comment. We added this phrase to explain in few words the original principle of the generator: "*This specific generator condenses heated vapours of diethyhexyl sebacate (DEHS) homogeneously on thin particles of sodium chloride, referred to as nuclei, to form monodispersed liquid particles. The size of these particles ranges from 1 to 8 µm, depending on the selected generation conditions.*"

RC2: "Line 198: What is the reason for the large gap between 0.15 µm and 2 µm particles? More sizes in this range would have allowed to gather some more information on the d50."

- You're absolutely correct, and you've highlighted a limitation in the generation process using glycerol and fluorescein. We attempted to generate a monodispersed aerosol of Glycerol with an aerodynamic diameter of 1µm and a GSD below 1.2, which is the criteria for an aerosol to be considered "monodisperse". Despite our best efforts to optimize the generation parameters, we were unable to achieve conditions that allowed the GSD to fall below 1.2. It's clear that the CMAG is optimized for use with DEHS. Any alteration in the liquid compound used for condensation on the nuclei impacts the generator parameters and the quality of the aerosol's monodispersion. This is why there are no experimental data points at 1µm, due to the lack of monodispersion in the generated aerosol. The use of a DMA or AAC could potentially meet the monodispersion criteria, but unfortunately, such instruments were not available during the experimental sequence.

RC2: "*Line 203-211: This paragraph can be shortened, because for spherical particles, the mobility diameter and the volume equivalent diameter are equal. In fact, this also what you describe, but with much too many words*."

- It's absolutely true. Only the simplified equation d_(ae)= d_m ((Cu(d_m) ρ_(p))/(Cu(d_ae) ρ_(0)))^(1/2) is now provided.

RC2: "*Lines 210, 214 and 217: The unit of the density is missing.*"

- Thank you for this comment. The unit of density is now mentioned throughout the text.

RC2: *"Lines 228 ff/Table 3: Why does the aerosol with MMAD = 0.16 µm have such a large GSD? Is that probably an artefact from the FMPS?*"

- No, we explain this large GSD by the fact that, for this specific diameter only, the particles measured are actually nuclei generated by removing the glycerol from the CMAG. Thus, without glycerol condensation on their surfaces, their diameters couldn't be homogenized. In summary, we typically measured the particle size distribution of nuclei generated before condensing glycerol on them to produce micron-monodispersed particles.

RC2: "*Lines 242 ff/Table 2: At this point it is not clear to me, why you investigated the effect of different dimensions, i.e. whether these were deliberately chosen or a result from manufacturing imprecision. Only later, I realized that the latter seems to be the case. This should be decribed more clearly here*."

- We appreciate the reviewer's feedback on that point. We acknowledge that the rationale behind this investigation was not explicitly stated in the manuscript. We have now revised the relevant section to provide a clearer explanation. The dimensions of the SADS prototypes were indeed a result of manufacturing imprecision, as detailed in the revised text. We have also provided additional context regarding the measurement of these dimensions using scanning electron microscopy (SEM) for accuracy.

RC2: "*Line 343: The reference should be for Figure 5, not 6*"

- Thank you for the comment. The mistake is now corrected.

RC2: "*Line 349 ff/Figure 5: How do you defined low, medium and high air[EB1] tightness?*"

- These levels were defined arbitrarily for classification at the end of the Mat&Met section (section 3.8 Leak evaluation") but we used a different name instead in the previous version of the article(leakage rate). That was probably misleading. The text of section 3.8 has been modified and we now use air tightness everywhere.

RC2: "*Line 385/386: The ratios of SADS 1 (1.27) and SADS 2 (1.29) are outside of this range. However, from Figure 5 it seems that SADS 1 has rather low wall deposition. Any idea why?*"

- Thank for pointing out this imprecision in the text. In the paper of Marple and Chien (1980), they state "it is recommended that the value of D1/D0 be kept less than 1.49, and preferably near 1.33". So, we correct our text accordingly by "All ratios are, however, below 1.49 as recommended by Marple & Chien, 1980a.". You once again highlight the fact that SADS 1 exhibited lower wall deposition compared to SADS 3, despite having the same nozzle ratio difference compared to the reference SADS R. Upon closer examination of the nozzle diameters, we observed that the diameter of the acceleration nozzle in SADS 3 is smaller than that of SADS 1. The theoretical developments conducted during this study indicate that a smaller diameter of the acceleration nozzle results in higher wall deposition. Therefore, we believe that the reduced diameter in SADS 3 is the reason for increased wall deposition in the acceleration nozzle, leading to a slight decrease in mass transmission efficiency.

RC2: "*Line 290/Figure 7: From Figure 5, I thought that ηp for MMAD=3.11 µm was ~90%. Doesn't this contradict the results in this figure?*"

- The two figures referenced do not depict the same set of experimental data. As emphasized throughout the paper, the lack of reproducibility is underscored here. Specifically, a notable disparity is observed when comparing the results for SADS 1 between the high air tightness condition in Figure 5 and the data for SADS 1 at 3µm in Figure 7. Conversely, the data for SADS 2 and SADS R are comparable in both figures.

RC2: "*Line 433-437: If the CFD results don't show the high wall deposition, could this be a result of the (wrongly) chosen turbulence model?*"

- Of course the whole CFD model present limitations. However, the turbulence model used actually overpredicts wall deposition, as explained in Belut et al 2022. This is due to the fact that it assumes isotropic turbulence near the wall, however turbulence is highly anisotropic there. Consequently the predicted wall normal turbulent dispersion of particles is exagerated and so is particles deposition (see also Mehel et al 2010 [1]). These points are however largely developped in Belut et al. 2022, That's why we prefer to refer readers to this article, in section 3.9. To tentatively help readers identify at a glance the limitations of the simulations as explained in Belut et al 2022, we propose to summarize these limitations with the following sentence, again at the end of secton 3.9 :
  "Following Belut et al. (2022), simulation results are realistic, within the calculated uncertainties, unless one of the following occurs: 1) the SADS walls are not smooth, 2) there is a difference between the actual and simulated geometry, 3) residual turbulence exists at the SADS inlet (with a Kolmogorov timescale much greater than the aerodynamic response time of the particles, which does not correspond to normal ambient conditions)."
  [1] Mehel, A., Tanière, A., Oesterlé, B., & Fontaine, J.-R. (2010). The influence of an

anisotropic Langevin dispersion model on the prediction of micro- and nanoparticle deposition in wall-bounded turbulent flows. Journal of Aerosol Science, 41(8), 729–744. https://doi.org/10.1016/j.jaerosci.2010.04.011

RC2: “Line 455: I believe this should say “…maintaining axial alignment..:””

- Thank you for the comment. The mistake was corrected.

RC2: “*Lines 467/471: what does H7/h6 and H8/f7 mean?*”

- These tags are international standards to describe mechanical backlashes, it is specified in the text : “Following ISO system of limits and fits, this corresponds to a H7/h6 clearance fit (location fit), whence a possible axial backlash in the range 0 to 0.041 mm is deduced.”**.**

RC2: “Line 477/Figure 11: the font size of the legend is very small; the symbol ηd cannot be used for the mass transmission efficiency and the particle deposition ratio!”

- ηd was defined as a "particle deposition ratio." Therefore, the second graph, placed on the right side, represents the particle deposition ratio for different misalignment distances. We believe that the font size was too small, which could have led to confusion between ηp used in the left graph of the figure and ηd in the right part of the figure. Now, the graphs have been separated into Figure 12a and 12b to be enlarged and made more readable.