

Interactive comment on “Application of a Hygroscopicity Tandem Differential Mobility Analyzer for characterizing PM Emissions in exhaust plumes from an Aircraft Engine burning Conventional and Alternative fuels” by Max B. Trueblood et al.

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Anonymous Referee #2

We thank the referee for a very thorough review of our manuscript.

The referee's comments on various topics were very valuable and we believe that ad-

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addressing these issues considerably improves the manuscript.

–reviewer's comments (in italic typeset, blue font). –a point-by-point response (in regular typeset, black font)

The paper consists of two main parts: 1) quality control of a hygroscopicity tandem differential mobility analyser (H-TDMA) and 2) experimental data on an aircraft engine. The H-TDMA is designed for fast response and stable humidity conditions. The quality control is done with care and shows a well functioning system. The experimental data gives important information. There are, however, some things that are unclear and I recommend publication after major revision.

General comments and questions:

RC#1 The authors define the hygroscopicity parameter soluble mass fraction (SMF). This value depends on the knowledge or assumptions of the chemical character of the soluble material. In the literature, several parameters have been used for hygroscopicity. κ defined by Petters and Kreidenweis (2007 and the discussion in ACPD) being the one mostly used today. But before that ε representing the soluble fraction under assumptions of the chemical composition was used. ε was abandoned, due to risks of misunderstanding. Why are you choosing to use a new parameter, similar to the one earlier abandoned?

RESPONSE #1: That entire section has been deleted.

RC#2 I have some comments on section 3 and the equations. a) Shouldn't it be $(Xw3 - Xd3)$ in eq. 1? It comes from the expression of the amount of water, I think. As far as I can see this has consequence for eq.8 and 9.

RESPONSE #2A: No, it is correct as it stands. Please refer to Eq. (6-33), p146, Pruppacher and Klett, (1978).

Eq. 1 is only valid for SR close to 1 (see e.g. Pruppacher and Klett, page 173 in the 1997 edition). Have you analysed the errors made at the SR values relevant here

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(0.85-0.99 according to line 27 on page 4)

RESPONSE #2B: We have analyzed the error brought about by this and have discussed this in the paper.

Even though the equations 2 and 3 are often expressed as they are here, I think it is unfortunate to give constants that actually have a unit, without expressing the unit. E.g. the constant in eq. 3 (4.3) includes the values of w and M_w as well as π . The value is 4.3 only if w is given in kg/litre and M_w in g/mole, and these are not the units in the SI system. This has been corrected in the manuscript now.

The constant 1000 in eq. 7 comes from not using SI units for M_s . Also, the result of this is that eq 9 is given in terms of π in some parts and with “combined” constants in other parts. As far as I can see, it would have been possible to simplify it, if the full expression of eq. 3 would have been used.

RESPONSE #2: This has been corrected in the manuscript.

RC#3 A more general comment to the calculations and the equations. Soot particles are in general agglomerates of many primary particles and their volume equivalent diameters are in general smaller than the mobility diameters. The particles can thus gain some secondary aerosol mass and water without increasing their mobility diameter. GF determined by H-TDMA systems can thus be below 1. I recommend that you take this into account or at least discuss it as a source of error.

RESPONSE #3: Gysel et al., (2007) stated that 15% of any given combustion particle is not soot. Thus we reduced the diameter X_d to $1/1.15=0.87$ of what the DMA1 provided.

RC#4 Small changes in GF for values close to one can have large influence on the cloud forming ability of the particles. Can you specify for example the lowest GF that is significantly larger than 1? Or the uncertainty in small GFs in general?

RESPONSE #4: We have included the following sentences in the revised manuscript: “The uncertainty in GF was 9% particles with diameter ~ 10 nm, and 3% for the larger

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diameters (26 nm). The uncertainty in κ was 7% and 2% for particles with diameter ~ 10 nm and ~ 26 nm, respectively.”

RC#5 P.8 l.31-32: It says: “ the SR-calc for the largest two or three particle diameters was computed and an average was obtained”. Does this mean that the theoretical curves in figure 2 are fitted to the largest sizes?

RESPONSE #5: That is correct.

RC#6 P.11 Fig.3 Are you sure that this is an effect of slow growth, and not of artefacts due to mismatch between DMA voltage and CPC counting? Have you tested with really slow voltage scans? Or scanning both up and down?

RESPONSE #6: Great care was taken to determine the lag time between when a voltage was imposed on the central rod of DMA2 and when particles selected by that voltage arrived at the CPC. Such a calibration took several days and we are confident of our result.

RC#7 Are there any ways to control the SR or do you have to work with the SR you get? Why are you working with such a high SR? I think it would be good to motivate this in the paper.

RESPONSE #7: The MST H-TDMA was purposely designed to take samples that do not last long and to operate in environments where the ambient conditions (say temperature) may change significantly and abruptly. It makes no sense to try to do a humidigram on a sample that is only present for perhaps 60 sec. Thus we opted to design an instrument that used only one SR, but held that SR very constant.

Other values of SR might be obtained by mixing air from the HUM and very dry air for both the polydisperse and the sheath air, but that would require more controls than we wanted to do. That might be an interesting path to pursue later on. It would allow longer operating times before the wetting tubes dried out.

Thus, if one can only have one SR condition, we deemed it best to use the condition

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that was most easily obtained and was the most stable.

RC#8 You have chosen to work with an aerosol to flow ratio of 3/15. A lower aerosol flow would increase the resolution and decrease the problem caused by a varying $dN/d\log D_p$. Could you discuss this in more detail?

RESPONSE #8: A lower Q_p and higher Q_{s2} would, indeed, provide higher resolution. However, a lower Q_p would also decrease the concentration seen by the CPC. A higher Q_{s2} would also increase the resolution, but again it would decrease the concentration seen by the CPC. Generally speaking, the higher the resolution, the lower is the concentration seen by the CPC. As mentioned in the text, the concentrations were already somewhat low sometimes, so we did not want to exacerbate that situation. Also, the wetting tubes dry out sooner for higher Q_{s2} flow rates. We felt that we had an optimal outcome here.

Details:

RC#9 P.1 I. 31 Spell out all abbreviations, e.g. UHC.

RESPONSE #9: This has been done and a list of abbreviations have been provided at the end.

RC#10 P.3 I. 8-9 Have you tested if the charger is strong enough to neutralize the aerosol?

RESPONSE #10: The bipolar charger is capable of housing 1 to 4 units of Po-210. Each unit has a strength of 500 μCi . That is 500 to 2,000 μCi . Po-210 is an Alpha emitter, which makes it considerably better at charging aerosols than a Beta emitter (Kr-85). This is because the specific ionization (number of ions created per centimeter of travel) of Alpha particles is much greater than that of Beta particles. Since the concentration of the aircraft engine particles was rather low by the time the plume reached the 143m sampling location, we are confident that the bipolar charger was strong enough.

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RC#11 P.3 I.28-29 Have you made sure that the whole cooling volume is cooled equally effective and that there are no "pockets" of water that is not circulated?

RESPONSE #11: The volume of the water bath surrounding DMA2 was approximately 12 L. The flow rate through the bath was approximately 5 L/min. Thus there was a complete water exchange every two minutes, a fairly short time. Furthermore, we point out that the SR was determined by a self-calibration using challenge aerosols of pure chemicals. So even if there were non circulated water pocket, the effective SR was determined.

RC#12 P.3 I.9 Are the 104 increments equally separated on a linear or a logarithmic scale or separated in another way?

RESPONSE #12: The logarithm of the voltage vs. time is linear.

RC#13 P.4 I.21 Performing HV2 sweeps on 12 different particle sizes in 9 minutes is very impressive!

RESPONSE #13: The MST H-TDMA was designed to study samples that were available for a short period of time (such as the plume from an aircraft landing or taking off) and/or a sample from a source that is very expensive to operate. Thus we tried to maximize the amount of data. Thank you for the compliment.

RC#14 P.4 I.27 The range in SR given is wide. Why is that?

RESPONSE #14: When this instrument was first deployed in the field, it was not as well insulated as it is now. Thus the SR-calc from using challenge aerosols of pure chemicals was somewhat lower, say 0.85 to 0.91. Now that it is better insulated the SR-calc values are typically 0.97.

RC#15 P.6 Fig.1 Make sure that all symbols are defined. For example P1 and P2.

RESPONSE #15: P1 and P2 are now defined within the schematic.

RC#16 P.6 I.11 There is an extra "nm" in the beginning of the raw.

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RESPONSE #16: This has been corrected.

RC#17 P.9 I. 13 Was the diameter 13,49 nm confirmed experimentally?

RESPONSE #17: No, it was not. But since the original diameter is computed using trustworthy equipment and we corrected that with theory, the 13.49 nm should be trustworthy.

RC#18 P.9 Fig.2 Consider the precision in the SR values. Also, please describe if the SR values are SR_{calc} or determined from the dew point sensor.

RESPONSE #18: The Reviewer must be referring to Fig 4. This is the plot of SR_{calc} vs Elapsed Time over a 240 min span. The ordinate is SR_{calc}. The uncertainty in SR_{calc} was stated in the figure caption as 0.008.

RC#19 P.13 Fig.5 There seem to be a drop in SR over the period presented. Could you quantify this drop and expand the SR scale to make it more sensitive.

RESPONSE #19: This drop seems to be about 6 parts in 986, or approximately 0.6 parts in 100, or approximately 0.5% in a four hour period.

RC#20 P.15 I.2 Why do you only present data from the 143 m location?

RESPONSE #20: The H-TDMA system was in a small trailer located 143 m downstream of the engine exit. This distance was deemed necessary to allow time for the hybrid particles to form from the insoluble cores and vapors of soluble species, such as H₂SO₄. Data from the other locations has been previously published in the literature (Moore et al., 2015). Only data acquired at the 143 m location are presented and discussed here to investigate the hygroscopic properties of the evolving plume.

RC#21 P.15 I.8-11 Did you see any bimodal GF distributions? I am especially thinking of the cases when the contribution from the engine was relatively small. Would it be possible to distinguish the engine particles from the ambient ones by their growth factors?

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RESPONSE #21: We do not observe any bimodal GF distributions.

RC#22 P.15 I.7 I guess you mean that the GF is close to 1 and not to 0? And in my opinion the GFs are not close to 1, they rather seem to be 1.2. What could be the reason for this? It makes a large difference for their cloud forming ability.

RESPONSE #22: It should be noted that the growth factors never go to zero, even for the FT fuel, but rather to about 1.15. This may well be the result of the fact that the insoluble core is porous. Thus even if there is not a spherical shell of H₂SO₄ around it, there is probably H₂SO₄ in the pores.

RC#23 P.16 I.11 Did you apply the factor 0.869 for all data, independent of the soluble fraction, that is also for particles that probably has no or very little sulphuric acid?

RESPONSE #23: Yes, we did. That may well be the reason that even the FT fuel showed a GF of 1.15 or so. Also, see the response to RC#22.

RC#24 Fig. 7-11 Specify that the SMF assumes that the soluble material is sulphuric acid (if this is the case). Also for the low sulphur fuels.

RESPONSE #24: All references to SMF have been removed from the manuscript and replaced with Kappa based on the reviewer comment.

RC#25 The figures in general: Please provide error bars. The quality control should be able to result in error bars.

RESPONSE #25: Adding error bars to the plots would make them cluttered. We have instead included the uncertainty in the GF and Kappa values in the text.

Reference:

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, 7, 1961-1971, <https://doi.org/10.5194/acp-7-1961-2007>, 2007.

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