

## ***Interactive comment on “Intercomparison study of six HTDMAs: results and general recommendations for HTDMA operation” by J. Duplissy et al.***

**J. Duplissy et al.**

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### Responses to Referee Comments

First, we would like to thank both referees for their constructive comments. The revised manuscript has been prepared including the reviewer's suggestions and comments. A point by point response to their concerns can be found (*italics*) below.

### Responses to Anonymous Referee 1 Comments

Page 128, line 26ff Should read: ...and the hygroscopicity tandem differential mobility analyser technique (HTDMA; Liu et al., 1978; Swietlicki et al., 2008).

*Sentence corrected according to the referee's suggestion.*

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Page 130, line 14f Should read: The sizing of the aerosol after humidification can be done in two alternative ways.

*Sentence corrected according to the referee's suggestion.*

Page 132, line 4 f In HTDMA the aerosol should not pass a pump before measured. Mostly one protects the pumps (in the sheath air cycle) by particle filters. Insofar aerosols inside a pump at higher temperatures here seems to be an unimportant aspect.

*The sentence on Page 132, line 4 f "Exposure of the aerosol to higher temperatures inside a pump may also alter its chemical properties through changes in the gas phase / aerosol partitioning." has been removed.*

Page 132, line 10 What is a recalculating air stream ? Should read: recirculating air stream ?

*We thank the referee for pointing out this mistake.*

Page 132, line 11 The phrase only possible should be replaced by only meaningful.

*This has been corrected.*

Page 132, line 23 and other places in the manuscript I suggest not to decline abbreviations like DMA2s

*Declination of abbreviations in this sentence and at all other instances has been removed.*

Page 133, line 7f The open sheath air systems does not apply to the described instruments. There must be good reasons, why all use closed sheath air cycles. Please, describe the advantage here.

*Some of advantages of the closed loop setup are listed at the beginning of Section 2.2.1. The following sentence has been added: "Furthermore it is easier to control*

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*sheath and excess air in a closed loop setup."*

Page 133, line 19ff Alternatively, Nafion dryers ...This sentence is difficult to understand, please rephrase it.

*The sentence has been replaced by: "Alternatively, Nafion™ dryers can be used, which require an extra source of dry air or an additional vacuum pump."*

Page 133, line 26 New paragraph after (Cruz and Pandis, 2000).

Done.

Page 134, line 3 Should read: ...are summarized in Table 1 and Fig. 1.

Done.

Page 134, line 5 What exactly is meant by growth factor equilibration ? growth factor observed at equilibrium ?

*This sentence has been replaced by: "A residence time sufficiently long to reach the equilibrium growth factor at an RH virtually equal to the RH in DMA2 is required such that no significant growth occurs during sizing in DMA2. "*

Page 134, line 21 The loss of semivolatle dicarboxylic acids in HTDMA was discussed in Koehler et al. ACP, 2006 (and references therein)

*Reference to Koehler et al. ACP, 2006 has been added.*

Page 135, line 9 Should read: ...(Johnson et al. 2008)

Done.

Page 135, line 13f This sentence is difficult to read, please rephrase.

*The sentence has been replaced by: "The RH in DMA2 determines the GF which is measured by a HTDMA given that the residence time at this RH is sufficiently long to reach equilibrium."*

Page 135, line 19f Should read ...thus illustrating the large uncertainty of GF imposed through the uncertainty in/of the RH.

Done.

Page 135, line 24f For this study the temperature probes... Really, only for this study ? How was T measured in HTDMA5 and 6 ?

*"For this study" has been removed from the sentence. For HTDMA5 and 6 the temperature was measured with the RH sensor (Humicap sensor from Vaisala, as said in the text). Figure1 has been updated by adding the temperature sensors for HTDMA5 and 6. The paragraph has been rephrased by: "HTDMA5 and 6 used capacitive RH sensors (Humicap Sensors from Vaisala) for direct RH and temperature measurements, which are less expensive but also less accurate than dew point hygrometers. The HTDMA1 to 4 measured the RH indirectly through accurate temperature and dew point measurements using a PT100 resistance temperature detector (RTD) and a dew point mirror (DPM), respectively. The temperature probes were placed in the water bath (HTDMA4), attached to the outer wall of DMA2 (HTDMA1), inside DMA2 (HTDMA3), or in the excess air flow (HTDMA2)."*

Page 136, line 5f The temperature in DMA2 must be measured mandatory. Therefore in case of capacitive RH measurements only one extra temperature measurement is needed together with the RH measurement e.g. in the excess air.

*The sentence "In contrast, capacitive RH measurements have to be done directly in DMA2 or two additional temperature measurements are needed for correction of temperature changes." has been replaced by: "A disadvantage of using a capacitive sensor is that the RH measurement has to be done in the sizing column of DMA2. Otherwise additional temperature probes are required to correct the measured RH for potential temperature differences."*

Page 136, line 8 Should read:...measure at RH < 90

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*Done.*

Page 137, line 3  $\alpha$ -Pinene (capital P at beginning sentence)

*Done.*

Page 138, line 4 Should read: Eventually DMA2 will stabilize at the same RH as DMA1 and hence the true GF of aerosol particles passing the HTDMA should be unity.

*Done.*

Page 138, line 8 Should read better: ...growth factors as proposed by Gysel et al. (2008).

*Done. (The reference was also updated to Gysel et al., 2009).*

Page 138, line 10 Should read: Additionally, the final RH in DMA2 shows whether the RH in DMA1 is sufficiently low, when operated in dry mode.

*Corrected as suggested.*

Page 139, line 15ff Rood et al., Nature 1989 stated early the ubiquitous nature of metastable ambient aerosols

*This reference has been added.*

Page 139, line 24f Should read: A validation at a single RH is only a weak test because of possible compensating effects from DMA performance, shape factors, restructuring, RH bias or impurities cannot be always excluded. Otherwise I start to lose trust in HTDMA measurements in general.

*Done.*

Page 140, line 13 Should read: ...at a nominally constant RH the actual RH in DMA2...

*Done.*

Page 140, line 14ff I suggest to show Gysels equation here. I understand that it is not

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a correction, but a recalculation to the nominal RH as long as deviations are smaller than +/-2

*The paragraph has been change by: "During measurements the RH in DMA2 varied about the target value. This caused concurrent variability of measured GFs. An empirical equation has been applied to recalculate measured GFs to their corresponding value at the target RH, thus minimizing this unwanted variability. Only measurements made at RH within a band of +/-2% around the target RH were considered and corrected according to the approach described in Gysel et al. (2009). Corrections of measurements to the nominal target RH are generally recommended in order to improve comparability of results from different studies. "*

*We believe that it is not necessary to repeat the equation shown in Gysel et al. (2009).*

Page 140, line 25ff Secondly... This is a complicated sentence for the relative simple fact that neglecting doubly (multiply) charged particles leads to an underestimation of the growth factor because they carry less than the double (triple etc.) of the mass due to slip (non-Stokes behavior). Please, rephrase and explain simpler.

*The following sentence "Secondly, the measured single charge equivalent mobility growth factor is smaller than the physical diameter growth factor for aerosol particles carrying multiple charges due to the nonlinearity of the Cunningham slip correction." has been modified to "Secondly, converting measured electrical mobility growth factors into physical diameter growth factors depends on the number of charges on a particle. Assuming a single charge for multiply charge particles results in an underestimation of the physical diameter growth factor due to the nonlinearity of the Cunningham slip correction. "*

Page 141, line 5 There is not much extra gain in Fig. 2 panel b. It is not even mentioned in the captions of Figure 2 at all. You should skip panel 2b.

*We agree and we keep only panel b. The legend of the figure has been changed to:*

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*"Figure 2 shows an example of ambient aerosol number size distribution (red shading). The blue line indicates the raw counts recorded by the CPC as a function of electrical mobility diameter set at the DMA. The bottom panels show the proportion of aerosol particles carrying +1, +2, or +3 charges. In the example, the singly charged aerosol particles dominate (=80%) at set diameters bigger than 105nm and an accurate HTDMA analysis is possible. Between diameter of  $D_0 \sim 60\text{nm}$  and  $D_0 \sim 105\text{nm}$  the selected physical diameter is not well defined because  $\sim 40\%$  of the aerosol particles are doubly charged, and data analysis is hampered. At  $D_0 \sim 60\text{nm}$  doubly charged particles dominate (=60%)."*

*The text on page 141 lines 5 to 11 has been adapted: "For illustration, Figure 2 shows an example of number size distribution, as observed during field measurements, along with the calculated size dependent number fractions of singly, doubly and triply charged aerosol particles. Often the singly charged aerosol particles dominate (>80%) and the HTDMA data analysis is straight forward and not biased by the multiply charged aerosol particles. Under certain conditions the number fraction of multiply charged aerosol particles can be significant, e.g. at  $D_0 = 60\text{nm}$  in Fig 2b."*

Page 142, line 21f Do you mean the accuracy at 90% RH ? If so state it clearly. Fig. 1 does not contain information of the accuracy of RH measurements.

*The sentence "All HTDMAs were able to hold the variability of the RH measured in DMA2 within the uncertainty imposed by the accuracy of this measurement (Table 1 and Fig. 1)." has been replaced by "All HTDMAs were able to hold the variability of the RH measured in DMA2 within +/- 2% of the target RH (Table 1)."*

Page 143, line 16 Why were HTDMA2 and 3 measuring something different. Please, you must explain!

*Sentence replaced by: "This test has been done with ammonium sulphate aerosol for HTDMA1 and 4-6 during the second campaign and with citric acid for HTDMA2 and 3 during the first campaign."*

Page 143, line 17 RH variations in DMA2 are undesirable though acceptable if they remain within limits. Is a soft statement. Please, make more precise statement, which limits.

*The sentence has been replaced by: "RH variations in DMA2 are undesirable though acceptable if they remain within +/- 2 % RH."*

The next sentence starting with However ... is also difficult to understand. Please rephrase.

*"However, this is only true if the GF measurements remain accurate despite the RH drifts imposed by the room temperature variations ." has been replaced by "However, this is only true if the RH measurement remains accurate despite the room temperature variations."*

Page 144, line 24 Since HTDMA 3 did something different, you may consider to skip this data in Fig. 4. The behavior at low RH is anyhow difficult to recognize.

*We think it is important to show the data from HTDMA3 since the accuracy has been affected by the change of the temperature even at low RH (green curve). All ordinates on the right-hand side have the same scaling in order to make the amplitudes of the green curves directly comparable for all panels...*

Page145, line 15f Should read better: wherein only data points with an RH gradient <1% during a scan were considered.

*The sentence "This included complete growth curves as shown in Fig. 5, whereas only data points with an RH gradient during a scan of  $\Delta RH < 1\%$  are considered." has been replaced by "This included complete growth curves as shown in Fig. 5, wherein only data points with an RH gradient < 1% during a scan were considered."*

Page 145, line 20 What other instruments do you refer to ? References or rephrase the statement.

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*We replaced the word "instruments" by "HTDMA".*

Page 146, line 12 You start to use 2ndDMA instead of the previously used DMA 2, please stay with DMA2.

*Done.*

Page 147, line 10 Should read: ...because the measurements were done at different RH in different studies. The performance of HTDMA for SOA can therefore be best assessed by simultaneous measurements. SOA generated at the same conditions should have the same properties.

*Done.*

Page 147, line 15 Do you have independent information on this ? It is a highly speculative and also dangerous statement. The difference in growth factors in the exp 1/2 and exp3 observed by HTDMA1 is quite significant. If it is really due to the bag, can we then still trust the older measurements? The higher growth factors in exp1/2 may be also due to wall effects, e.g. uptake of water soluble material evaporated from the walls (memory effects).

*This paragraph has been reformulated to: "SOA produced by photo-oxidation of  $\alpha$ -pinene was investigated during the two workshops. HTDMA1, the only instrument participating at both workshops, measured slightly different GFs (Figures 6 and 7). The SOA properties from the third experiment are not representative because a new smogchamber bag was used, which had not yet undergone the standard cleaning procedures. However, this does not compromise the suitability of the generated SOA for comparing the performance of different HTDMA instruments."*

Page 147, line 24 By not cycling through a series... This is difficult to follow, please rephrase positive.

*The sentences "The general sampling strategy employed was to measure just one dry size for several hours and then switch to a larger dry size. By selecting the largest dry*

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size at which there were sufficient statistics it was possible to measure that size for a longer time without the influence of multiply charged aerosol particles (as the mode of the size distribution grew). By not cycling through a series of different dry sizes one could not see clearly if there was any difference between them at a specific time in the experiment."

have been replaced by

*"The general sampling strategy employed was to measure just one dry size for several hours and then switch to a larger dry size. The upper and lower limit of possible dry size at a certain time are imposed by low particles concentrations and biases due to the influence of multiply charged particles respectively. Both size limits increase with time as the mean diameter of the size distribution increases. Always starting off with the largest possibly dry size made it possible to measure at a constant dry size for a longer time before changing to a larger dry size. This strategy was chosen to measure possible temporal trends of hygroscopic growth due to photochemical aging, while it is not suitable to determine whether or not the hygroscopic growth at a certain time is size dependent."*

Page 148, line 10 Should read: In experiment 3 (Fig. 7) HTDMA1 measured a slightly lower growth factor than in experiments 1 and 2. The difference measured by HTDMA1 between exp3 (GF = 1.14 - 1.22 over 5 hours) and Exp1,2 (GF =1.25 - 1.4 in 8 hours) seems to be but quite significant. What is the reason for that difference. Can emanating (oxidized) impurities from the new Teflon bag cause such a large decrease ?

*The sentence "In experiment 3 (Fig. 7) HTDMA1 measured..." has been deleted, because this statement has already been made above (p. 147, line 15). With the answer to the above comment (p. 147, line 15) we have already clarified that the SOA produced during experiment 3 is not considered to be representative.*

Page 148, line 13 If two instruments measure a difference in GF for the same SOA the reason must be in the instrument performance or construction and it cannot be in the

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SOA.

*The referee is right, the original statement was misleading. The paragraph has been reworded: "These experiments revealed significant discrepancies between the HTDMA measurements for SOA, whereas they agreed well for inorganic salts. This implies that the SOA has a particular property, which makes it much more difficult to obtain accurate growth factor measurements using a HTDMA. Based on the tests performed during these experiments no instrument design feature could be identified as an obvious reason for these discrepancies. Hypotheses for possible causes are discussed in Section 4.3."*

Page 150, line 20 The introducing sentence is too complicated, it should be rephrased.

*The sentence "Based on the results of these workshops, recommendations for building a new generation of HTDMAs for field operation at the EUSAAR supersites, as well as standardised calibration, quality assurance and data analysis procedures are provided in the following:" has been rephrased by "In the following we provide recommendations for design and construction of HTDMA instruments as well as for standardised calibration, operation, quality assurance and data analysis procedures, based on the results of these workshops. Several key points which are commonly acknowledged by experienced HTDMA users are also included for completeness."*

Page 153, line 5 With exception of the first bullet, these points are anyhow mandatory for any DMA or SMPS measurement. I don't see them as a special outcome of the two workshops ! That was known before, wasn't it ?

*Yes, we agree that this was known before, however it was not always applied. We believe that repetition of this basic recommendation is very important for unexperienced HTDMA users. This point has been made clear with the changes made according to the previous comment.*

Page 152, line 11 Should read better: Certified PSL spheres should be used to verify

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the sizing of the DMA. This should have been also mandatory for each DMA measurement !

*The bullet point "Certified PSL spheres can be used to verify the sizing of the DMAs" has been replaced by "Certified PSL spheres should be used to verify the sizing of the DMA" in order to make it clear that verification of the DMA sizing with PSLs SHOULD be made.*

Table 1 The use of DMA1, DMA2 and first, second DMA should be homogenized Some headers in column 1 do not start with capital letter: Water source, Sizing

*Done.*

Table 2 What exactly is the difference between no of measurements and number of points.

*This has been corrected. "Number of points" has been replaced by "number of measurements".*

Fig. 2 Total number is shown by blue lines. Panel b is not mentioned.

*As discussed previously panel a has been removed and panel b is now discussed. In the text "The black line..." has been replaced by "The blue line..."*

Fig, 4 You may consider to take out the data of HTDMA3.

*We prefer to keep the data of HTDMA3, see arguments above.*

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Interactive comment on Atmos. Meas. Tech. Discuss., 1, 127, 2008.

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