

Interactive comment on “Tandem configuration of differential mobility and centrifugal particle mass analyzers for investigating aerosol hygroscopic properties” by Sergey S. Vlasenko et al.

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The authors propose to use tandem DMA-HCPMA measurements to measure hygroscopic properties of particles. The paper includes a good discussion of the pros and cons of competing methodologies, and clearly explains conceptual advantages of the measurement method they describe. It is an appealing approach.

Nevertheless, I have two major concerns and some minor ones:

Major concerns:

(1) My major concern is the cavalier discussion of measurement technique fundamentals. Figure 2 shows particle number mass distributions with no explanation as to how

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they were obtained. In fact, given the relatively broad transfer function of instruments such as the CPMA, obtaining number mass distributions is not straightforward. The literature includes some pertinent information, which is not discussed.

Park et al. (Park et al. 2003) reported on measurements of aerosol mass distributions as a function of mobility diameter from DMA-APM measurements. Their method assumed that particles classified by the DMA had only a single mass (the "modal mass" in the language of the paper under review), which is only approximately true even for chemically homogeneous aerosols. Because the transfer function of a DMA is triangular, mobility-classified particles have a distribution of masses and that distribution affects APM (or CPMA) data. The transfer function of the APM (or CPMA) is even broader than that of the DMA, so at the voltage corresponding to the "modal mass" some particles of every size leaving the DMA penetrate through the APM (or CPMA). Furthermore, ambient aerosols of a given mobility size may include particles of that are chemically and morphologically distinct, which leads to multimodal mass distributions. These subtleties need to be acknowledged in a measurement techniques paper.

More recently, Rawat and coworkers (Rawat et al. 2016) developed an inversion algorithm for obtaining two dimensional number distributions (as a function of mobility diameter and mass) from DMA-APM measurements. Equation 2 of that paper shows the relationship between measured number concentrations downstream of the APM, and operating characteristics of the DMA-APM apparatus (flow rates, voltages, etc., which determine the DMA & APM transfer functions.) Extending their approach to DMA-CPMA data should be possible provided the CPMA transfer function is sufficiently well known. However, Vlasenko and coworkers do not discuss this conceptually important background. I suspect the number mass distributions shown in Figures 2 & 4 were obtained by assuming that the number mass distribution was constant at a given CPMA classifying voltage. Given the narrowness of the sampled aerosol distribution and the breadth of the CPMA transfer function, that is a not a good assumption.

Most previous DMA-APM (or CPMA) work has involved working with raw data: i.e.

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measurements of number concentration downstream of the CPMA as a function of CPMA classifying voltage (or equivalently, modal mass), and this is a valid approach. If the authors choose not to use a mathematically justified approach for inverting data to obtain number distributions with respect to mass, I would recommend that they stick to analysis of the raw data. This would involve revising figures 2 & 4 to show only N versus VCPMA, or equivalently, N versus modal mass. The figures 3 & 6 are based on the modal mass (i.e., the value that corresponds to the peak value in the N(VCPMA) measurements), so as far as I can tell the distribution functions are not required for the analyses that were done.

(2) While I this methodology is conceptually appealing, I do not believe the paper delivers on the abstract's promise: "The direct measurements of humidified particle mass allow avoiding complications that occur in the commonly used mobility-diameter-based HTDMA technique due to poorly defined particle morphology and density." It is clear from results of the paper that heating within the CPMA and the broad transfer function of the CPMA lead to complications that are at least as great as those that occur with the HTDMA. The abstract fails to provide a straightforward assessment of the proposed measurement technique's weaknesses. The abstract needs to be forthright about identifying those weaknesses.

Minor Concerns:

(1) Based on results presented in the paper, I think a strong case can be made that the HTDMA method is in principle better for measuring deliquescence and efflorescence thresholds. It is easier to operate a HTDMA under isothermal conditions.

(2) On p. 5 it is stated "... for AS and NaCl particles with initial mobility diameter $D_b=60$ nm and dry masses of 0.18 ± 0.01 fg and 0.21 ± 0.01 fg." I assume " ± 0.01 " corresponds to the estimated uncertainty in the modal mass. While the modal mass may be known with high certainty, the relatively broad transfer function of the CPMA ensures that the range of masses exiting the CPMA greatly exceed this value. The authors need to

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explain why the modal mass is the proper variable even though sampled mass distributions might have been multimodal (see point 3 below).

(3) The abscissas of Figures 2 & 4 are labeled "Particle Mass". I recommend they be relabelled "Modal Mass". The reader needs to understand that, in fact, particles covering a broad range of masses were present at each CPMA voltage. The importance of this is emphasized by the discussion on p. 6 "...the output aerosol in dehydration mode is a mixture of droplets and dry particles..". If the CPMA transfer function were sufficiently narrow, it would have been possible to distinguish between droplets and dry particles. It is also possible (but not guaranteed) that this could have been achieved if an inversion method similar to that discussed by Rawat et al. had been used to retrieve the true mass distribution. In any event, this phrase supports my argument that these plots do not show mass distributions and need to be replotted.

(4) I am confused by Figure 3. For AS, the blue "+" is labelled "non-prompt efflorescence" while for NaCl, the blue "+" is labelled "Non -prompt deliquescence". The text on p. 5 states "For both AS and NaCl particles, intermediate growth factors between dry and deliquesced particles were observed (Fig. 3 - blue crosses)." The text contradicts the figure label.

(5) I am not convinced that contact efflorescence explains the results and that this might be an approach for studying contact efflorescence (see Figure 5 and discussion on p. 6). Wouldn't it be possible to test this idea by carrying out measurements extending from high voltages, where all particles reach the inner rotating electrode, to low voltages, where all particles reach the outer rotating electrode? The proportion of particles undergoing contact efflorescence should be higher at the low or high voltages, right? Is there any evidence for this?

(6) Does the extent of non-prompt efflorescence and deliquescence change if measurements are carried out when the CPMA is first turned on (i.e., before frictional heating has had time to warm it up)?

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In summary, the proposed measurement methodology offers clear conceptual benefits over other methods such as the HTDMA for studying particle phase transitions and hygroscopicity. However, the measurements that are reported reveal limitations on measurement accuracy that may be difficult to overcome. Furthermore, I question the validity of Figures 2 & 4. Because the method, in principle, adds to what can be learned from other techniques, I feel it would merit publication after the authors respond to the points raised above.

Park, K., D. B. Kittelson and P. H. McMurry (2003). "A closure study of aerosol mass concentration measurements: comparison of values obtained with filters and by direct measurements of mass distributions." *Atmospheric Environment* 37(9-10): 1223-1230.
Rawat, V. K., D. Buckley, S. Kimoto, M.-H. Lee, N. Fukushima and C. J. Hogan Jr. (2016). "Two-dimensional size-mass distribution function inversion from differential mobility analyzer-aerosol particle mass analyzer (DMA-APM) measurements." *Journal of Aerosol Sci.* 92: 70-82.

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