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Interactive comment

Interactive comment on "Data inversion methods to determine sub-3 nm aerosol size distributions using the Particle Size Magnifier" *by* Runlong Cai et al.

Anonymous Referee #1

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General comments:

This work reports the accuracy and capability of four data inversion methods for analyzing the data obtained by the particle size magnifier (PSM). The authors discovered that among all the data inversion methods, the expectation-maximization (EM) algorithm provides the best agreement with the test aerosol size distributions both experimentally and computationally. However, all four data inversion methods generated false sub-3 nm particle concentrations when the aerosols were larger than 3 nm, because of the limited resolution of the PSM in controlling the saturator flow rate. Suggestions regarding the PSM operation and data inversion were also given based on the findings

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of this work. This work will be a very useful publication regarding the interpretation of the PSM data because the PSM is receiving wide applications in new particle formation studies. The reviewer recommends the publication of this manuscript after a few points regarding the resolution of the PSM and the data inversion methods are properly clarified.

Specific comments:

1. Resolution of the PSM: The reviewer understands that the resolution of the PSM should be defined based on the saturator flow rate instead of the particle size, since the relationship between the saturator flow rate and particle size are dependent on the chemical composition and charging state of the sub-3 nm particles. But it may still be helpful to translate the resolution in terms of particle size since this study does not consider the influence of the chemical composition and charging state. It was mentioned in the manuscript several times that the low resolution of the PSM led to the detection of false sub-3 nm particles even when the particles are above 3 nm. But one wonders how this "low resolution" (e.g. $\sim$ 1.0 in Page 11 Line 4) may relate to the resolution in terms of particle size. For example, in this study, does a resolution of $\sim$ 1.0 at 3.93 nm (based on the saturator flow rate) simply mean that particles of 3.93 nm can be detected by the PSM in the size bins between 3.93-1.97nm and 3.93+1.97 nm?

2. Stability of the non-negative least squares method: In this study, both the kernel function method and the H&A method used the non-negative least-squares method (probably the "lsqnonneg" function in MATLAB) to directly solve the particle size distributions. This function indeed can cause instabilities when the inversion matrix becomes complex. The reviewer wonders whether the authors could use the Twomey inversion algorithm to further refine the solution by using the results of the non-negative least-squares method as an initial guess. One can refer to Eqs. 3 and 4 of Markowski (1987) for further detailed calculation methods. In this way, the instability of the non-negative least squares method can be reduced. The smoothing algorithm could be disabled (neglecting Eqs. 6 and 7 of Markowski (1987)) if the authors are concerned

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with its adverse influence on data inversion. Since the Twomey inversion method involves the iteration of linear equations, the computational expense should be low as well.

Reference: Markowski, G. R. (1987). "Improving Twomey's algorithm for inversion of aerosol measurement data." Aerosol Sci. Technol. 7(2): 127-141.

3. The detection of false sub-3 nm particles: The reviewer is quite puzzled by the detection of false sub-3 nm particles when the test aerosols were above 3 nm. Take the stepwise method for an example, theoretically, according to Eq. 4, n_m would become 0 when the test aerosols were all above 3 nm. Even if we consider the limited resolution of the PSM for particles above 3 nm, the PSM should not report the detection of aerosols below 1.5 nm, which is shown in Figures 5 and 6. The reviewer wonders if the false detection of the sub-3 nm particles is related to the "error/uncertainty" in both the simulation and experiments, rather than the low resolution of the PSM. In addition, could the authors show the PSM-measured particle concentrations as a function of the saturator flow in Figure 6, similar to the one in Figure 5b, so that the error/uncertainty during the experiment could be evaluated?

Technical comments:

1. Page 3 Line 19: "... a regularization parameter... and the agreement with the PSM recorded data, ..." Was PSM data analyzed by using the Tikhonov regularization method?

2. Page 5 Line 17: "size ability" –> "sizing ability"?

3. Page 6 Line 3: "step-wising" –> "stepwise", same applies to the rest of the manuscript.

4. Eq. (4): Please check the unit of n_m. R should have a unit of cmˆ(-3), and the denominator is dimensionless. Also, regarding the efficiency terms in the denominator, should they be eta(s_i,d_p,max) and eta(s_i+1,d_p,max), because the calculation is

specific for the ith and (i+1)th saturator flow rates?

5. Please check equations (6) and (8)-(10), they have some format issues on my computer.

6. Eq. (10): According to the definition of the H&A method, should the matrix Q have a dimension of J»I? Otherwise, please include some important steps converting the matrix into a square matrix.

7. Page 9 Line 23: “No./cm3” –> “cmˆ(-3)”?

8. Page 9 Line 30: What was the approximate time needed for the measurement in the stepping mode and how stable was the wire generator?

9. Page 15 Line 13: “sable” –> “stable”?

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