

Review of the manuscript entitled “Multi-modal analysis of aerosol robotic network size distributions for remote sensing applications: dominant aerosol type cases” by M. Taylor, S. Kazadzis, and E. Gerasopoulos, with reference **no.: amtd-6-10571-2013**

The present paper deals with the development of two methods of fitting the Aerosol Volume Size Distribution (AVSD) as well as with their application to AVSDs of distinct aerosol types. Specifically, the OEV (Optimized Equivalent Volume) method is developed to optimize bi-lognormal fits of AVSDs used by AERONET, and the GMM (Gaussian Mixture Model) method is proposed for fitting the AVSD with multiple modes. Secondary, in order to apply these two methods to cases of different dominant aerosol types, authors propose an approach based on the synergy of AERONET data and GOCART model outputs, for aerosol categorizing and the selection of sites and days with distinct dominant aerosol types.

Accurate determination of aerosol optical/microphysical properties from remote sensing observations is essential for various scientific problems (e.g. climate and climate change studies, air quality issues, ...) where aerosols are involved. Thus, any effort aiming at developing new retrieval algorithms or/and improving the existing ones is of great importance.

In this framework, the submitted paper is interesting, well written and organized whereas the developed models are well documented and robust in terms of statistics. In overall, it can be published in the AMTD Journal after taking into account the following comments.

The results of the comparison between the OEV bi-lognormal fits and the AERONET bi-lognormal fit (Tables S1 and S3) discussed in section 4 reveal relatively large relative errors for the secondary microphysical parameters (r_f , σ_f , V_f , r_c , σ_c and V_c), especially for the dust and marine aerosol types. Based on this information, have the authors examined whether the differences between those parameters derived from the OEV method and the AERONET bi-lognormal fits, are statistically significant?

Note that for the GMM method, authors state in the discussion (section 5, page 10593, line 22) that they “*performed a test for a statistically-significant improvement in the fit with the addition of each additional mode*”.

In section 4 (pages 10589-10590), authors state: “*... its impact on the values of the secondary microphysical parameters is dramatic.*” and prove through the estimated relative errors, that this is particularly true for dust and marine aerosols. I am wondering how feasible is for the authors to give an estimate on the effects of the proposed models on the “final” products such as the AOD of fine and coarse fraction and others.

It is obvious that the application of the proposed approaches and especially of the GMM method, brought improvements, both qualitative and quantitative (in terms of statistical measures), in AVSD fits compared to the reconstructed bi-lognormal AERONET ones. Though authors in the 2nd part of the section 4 and throughout the section 5 present the performance of both developed methods for all considered aerosol type, at the end I miss a clear conclusion or suggestion on which of them is appropriate for each case. This is not valid for the case of marine aerosols where it is clearly stated that only the GMM 3-mode model reproduce accurately the AVSD. For instance, even in the cases of urban and biomass burning aerosols, the best fit is again the GMM tri-modal. However, authors by invoking the physical significance of secondary peaks suggest that they can be approximated by bi-lognormal fits. So, is in those cases the OEV method the most appropriate or we can stay in the AERONET fits? Finally, for the dust case, I feel that an advantage is given to the OEV approach over the GMM without being so clear to me why.

The present work focuses on the presentation and description of the two new methods, while in terms of validation the proposed models are applied to 4 single cases of dominant aerosol types. It would be helpful to extent the application-validation to more cases so as to generalize the derived conclusions. Such investigation could give answers to my previous comment.

In the concluding section, I think that authors could reduce its length avoiding repetitions or information that is not really a conclusion (e.g. that statement “*it is possible to perform sensitivity analysis of the dependence of secondary microphysical parameters on (a) r_s and (b) the aerosol load (as measured by the AOD as a proxy),*”) and add a few sentences addressing the following issues:

- Whether authors intend to extend the application of their methods to other aerosol dominant cases and sites.
- What is the potential of those methods for a wider applicability to cases where various aerosol types coexist? In the beginning of the concluding section there is a relative reference to the OEV method. It would be interesting to give more information and include the potential of the GMM method too.
- In the last paragraph, authors could mention how feasible (easy and immediate) is the implementation of the proposed models to existing operational retrieval algorithms.

In section 3.2 (2nd paragraph, lines 16 – 28), authors write: “... R^2 is much more sensitive to changes in r_s and reveals a peak value of 0.893 at $r_s = 0.286 \mu\text{m}$.”, also indicated in figure 2. However, following in the same paragraph as well as in the next one, the value of $0.315 \mu\text{m}$ with $R^2 = 0.894$ is used for the r_s corresponding to max (R^2). Obviously the correct value is the one used ($0.315 \mu\text{m}$) and the sentence just refers to figure 2. Though next in the results section authors use the term “*the tabulated entry closest to the optimal OEV value ...*” to distinguish the estimated optimal r_s value than the one appearing in tables, it should be more clear a concise to make the necessary corrections. Authors could even replace in figure 2 the fit corresponding to $r_s = 0.286 \mu\text{m}$ with the one of $r_s = 0.315 \mu\text{m}$. If authors could not illustrate the actual optimal r_s values and the related secondary parameters in figures and tables, they could add in the paper a table similar to the table S3 with less information, namely the AERONET bi- lognormal fit and the best fit suggested by the two methods OEV and GMM.

While in section 3.1 authors explain why they use a large number of interpolation points, in section 3.2 they do not justify the choice of 2200 (2198 plus the two end-points) equidistant logarithmically-spaced radial bins against for instance, the maximum 2816 points. Then, in section 5 (page 10593, lines 16-20) they write “*The use of iterated nonlinear least squares to obtain the microphysical coefficients was very efficient – although it was necessary to interpolate the AVSD with a 100-fold increase in the number of points (from 22 bins to 2200 bins) so as to avoid numerical instability (i.e. so that the propagated errors of the fit were stable at the 95% level of confidence).*”, please give this explanation clearly in the appropriate section (methodology presentation). Do they converge to this number after test? Why not the 2816 points? Does it make any difference?

A technical comment: text in figures and especially the axis titles are illegible. Authors should improve the quality and enlarge the font.