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Interactive comment on “Multi-modal analysis of aerosol robotic network size distributions for remote sensing applications: dominant aerosol type cases” *by* M. Taylor et al.

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We thank the reviewer for their detailed review of the manuscript and for their support:

“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 AVDS 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“.

Below, we respond to each point raised by the reviewer in detail:

“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 (rf, σf, Vf, rc, σc and Vc), 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.”

We thank the referee for a very helpful suggestion here. While statistical hypothesis testing is at the core of the GMM method (as the referee point out), we agree that it is helpful also in assessing the impact of the sensitivity analysis used in the OEV. We performed a 2-tail paired t-test with the AERONET value of each parameter being kept constant over the range of rs used in the OEV method. We obtained the following results at the 95% confidence level ($p<0.025$ for a statistically-significant difference) shown in Fig. 1 accompanying this reply. We have added this as Table 3 in the manuscript and have added the following paragraph in the text at the end of the paragraph on page 10590 (line 20):

"In order to assess whether or not the changes in the secondary microphysical parameters arising from application of the OEV method are statistically-significant or not, we performed a 2-tail paired t-test on the values arising from application of the OEV method with the value of each parameter as provided by AERONET. The test was performed at the 95% level of confidence whereby a value of $p < 0.025$ reports a statistically-significant difference. The results of performing this test for each of the 4 dominant aerosol types is presented in Table 3 which shows that there is a statistical difference between fitting the AVSD with AERONET's reconstructed bi-lognormal and the OEV bi-lognormal only in the case of dust and marine (sea salt) Vf, Vc and rf - reinforcing our assertion that the AERONET fit of the AVSD of dust and marine-dominated aerosol is problematic. Application of the OEV method both 1) improves the fit in these two dominant aerosol cases and, 2) leads to significant differences in the values of the volume concentrations of the fine and coarse mode and also the fine mode geometric radius."

"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."

The referee's question is an important one. In our reply to Anonymous Reviewer #2 referring to this same point, we performed t-tests to ascertain which secondary microphysical parameters exhibit a statistically-significant difference when calculated with the OEV method as compared to the AERONET inversion. While the emphasis of our manuscript is on a new parameterization of the size distribution, it is of course per-

tinent to ask what impact this may have on "final" products. The inversion algorithm used by AERONET optimizes the size distribution and the spectral complex refractive index (taking also into account the proportion of spheroids in the atmospheric column) so as to match measurements of the intensity of direct radiation at several wavelength bands. It is not yet clear to us how the refinements in the fitting of the AVSD we suggest here translate to "final" products but a very rough estimate of their impact on, for example, the AOD at 1020nm can be obtained with reference to Fig. S2 and Tables S1 and S3 in the Supplement. We have added the following sentence explaining how to perform such an estimation in the manuscript following on directly from the addition at page 10590 line 20 resulting from the related point raised by Anonymous Reviewer #2:

"While our emphasis is on a new parameterization of the size distribution, it is of course pertinent to ask what impact the changes imparted on derived secondary microphysical parameters by the OEV method, may have on "final" products such as the AOD. In order to estimate this effect on, for example, the AOD at 1020nm we refer the reader to Fig. S2 in the Supplement. The lower 2 panels in Fig. S2 show the regression of Vc and Vf respectively on the AOD at 1020nm for the case of dominant marine aerosol at Lanai. For the OEV method the regressions have the form: Vf = 0.12 x AOD(1020) and Vc = 0.93 x AOD(1020). Inverting these linear relations, we find that for the fine mode, AOD(1020) $\approx$ 8.33 Vf and $\approx$ 1.08 Vc. To estimate the effect of application of the OEV method as compared to the results of fitting the AVSD with the AERONET reconstructed bi-lognormal, we refer to entries in Tables S1 and Tables S3 for marine aerosol where the AERONET separation point for this type of aerosol in Table S3 is 0.439μm. The entry in Table S1 closest to this separation point is rs = 0.587 which has a relative error (for AERONET-OEV) of $\approx$ -28% for Vf and $\approx$ +7% for Vc. We therefore expect that the AOD(1020) for the fine mode should be about 28% higher when using the OEV method than that predicted by AERONET and that the AOD(1020nm) for the coarse mode should be about 7% lower when using the OEV method than that predicted by AERONET".

“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”.

Thank you for this important comment. We have re-read sections 4 and 5 and agree with the reviewer that we have not made clear enough a couple of points: 1) the outcome of the comparison of the AERONET fit and the OEV method in the case of biomass burning and urban SU, and 2) the outcome of the comparison of the OEV and GMM methods in the case of dust. For the former, on page 10589 at line 17 we have added the following clarification:

“Table S2 in the Supplement shows that R2 = 0.983 for AERONET and R2 = 0.985 for the OEV method in the case of biomass burning, and that R2 = 0.982 for AERONET and R2 = 0.987 for the OEV method in the case of urban SU. While Fig. 6(b) and 6(c) suggest that there is almost no visual difference between the two fits for these two dominant aerosol cases, we will demonstrate later in this section that even the more noticeable difference in the mode separation point rs obtained for these two aerosol types (see Table S3 in the Supplement), do not translate into statistically-significant changes in derived secondary microphysical parameters. As such, it appears that the

AERONET fit is fine in these two cases.”

With regard to 2), on page 10591 at line 6 we have added the following short sentence:

“Table S2 in the Supplement reveals that, In the case of dominant dust aerosol, the GMM method with 2 modes provides an improvement in the goodness of fit: R2=0.995 as compared to the OEV method where R2=0.979. This improvement is also visually noticeable in Fig. 6(a).”

“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”.

Yes, we agree with the reviewer also here. In response to the suggestions made by Dr. Andrew Sayer, we constructed an ensemble of the 10 most dominant cases for each of the 4 aerosol types (kindly see our detailed response to this point you raise in our reply to Dr. Andrew Sayer below please). It is our intention to study the temporal evolution of a larger variety of cases including aerosol mixtures in a follow-up paper.

“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) rs 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 extent 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".

We thank you for these good suggestions. We re-read the conclusion and believe that the emphasis on the OEV and GMM is fairly evenly spread, and highlight the main findings of the work. Following the suggestion of the reviewer, we have added the following 2 paragraphs to the conclusion on page 10596 line 14:

"The methods described here can be readily implemented in existing operational retrieval algorithms by coding a post-processing module which reads in the AERONET retrieved AVSD, interpolates the reported values of dV/dlnr over a finer radial grid and then: 1) calculates secondary parameters using the equations presented in Appendix A by looping over a range of mode separation points (the OEV method), or (preferably) by performing nonlinear least squares fitting using multiple Gaussians in ln(r)-space and performing nested hypothesis testing with reference to the equations presented in Appendices B and C (the GMM method)".

"Having applied the OEV and GMM methods to dominant aerosol types cases, we are currently applying the methods to study the temporal evolution of AVSDs during atmospheric phenomena where ambient conditions are affected by the incursion of additional aerosol species that lead to abrupt changes in the chemical composition such as during volcanic eruptions, dense urban brown cloud episodes, desert dust storms and forest wildfire outbreaks, as well as modification of ambient aerosol conditions caused by the presence of fog and low-lying clouds."

As an indication of the light computational complexity of the task of potentially incorporating these methods into operational algorithms, all data loading, processing, interpolation, fitting and plot generation routines used to generate the results presented here were accomplished with just over 500 lines of MATLAB script.

"In section 3.2 (2nd paragraph, lines 16 – 28), authors write: "... R2 is much more sensitive to changes in rs and reveals a peak value of 0.893 at rs = 0.286 um.", also

indicated in figure 2. However, following in the same paragraph as well as in the next one, the value of 0.315 um with R2 = 0.894 is used for the rs corresponding to max (R2). Obviously the correct value is the one used (0.315 um) 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 rs 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 rs = 0.286 um with the one of rs = 0.315 um. If authors could not illustrate the actual optimal rs 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".

Thank you for your comment. On page 10582 lines 13-15 we attempted to make clear that in Fig. 2 we present the results of applying the sensitivity analysis to the radial range 0.1-7um. We believe that the source of confusion here is because we actually calculate the goodness of fit parameters over the full range of 2198 interpolated points (excluding the end points) used to step through the range of values of rs. To help the reader visualize the dependence on rs, we then extracted values at a coarser spatial scale and reported 16 values at equal steps in ln(r) across the radial range 0.1-7um. This coarser grid, however, does not specifically include the optimal point as the reviewer points out. As an illustration of the behaviour of the sensitivity analysis for the whole 2198 point spectrum, Fig. 2 accompanying this reply shows the R2 and SSE-curves as a function of rs together with the optimal values of rs as deduced with min(SSE) and max(R2) for the case of biomass burning used in the manuscript.

During our co-author discussion meeting prior to submission, we decided to cut this figure and explanation from the manuscript because we felt that it was too much information for the reader to digest while at the same time trying to retain their attention on the mechanism of the OEV method. We have addressed this in the revised manuscript by modifying the caption to Fig. 2 as follows from:

“Fig. 2. Sensitivity analysis of the equivalent volume bi-lognormal fit to the AERONET AVSD data with varying mode separation point rs for dominant marine (sea salt) aerosol at Lanai, Hawaii on the 21st of January, 2002. The grey squares are the values of the AERONET AVSD.”

to:

“Fig. 2. Sensitivity analysis. The equivalent volume bi-lognormal fit to the AERONET AVSD data obtained by varying the mode separation point rs over a coarse 16-point radial grid spanning the interval 0.1 to 7μm for dominant marine (sea salt) aerosol at Lanai, Hawaii on the 21st of January, 2002. Note that the optimal mode separation point (rs=0.303μm) described in Sect. 3.2 is obtained by applying the max(R2) and min(s) methods on the high resolution interpolation grid of 2198 points. The grey squares are the values of the AERONET AVSD.”

“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?”

We would like to thank you once again for your comment. On page 10580 lines 2-8, we outline briefly our rationale for interpolating the AVSD. However, yes, as you point out we did not justify the need for the high level of interpolation (2200 points). As for the sensitivity analysis above, we had prepared convergence plots like Fig. 3 accompanying this reply (for the case of dominant marine aerosol at Lanai). The upper panels show a rapid convergence of the interpolation volume concentrations with increasing number (N) of interpolation points for N >= 44. However, at such coarse radial resolution, the problem is that the standard error of the GMM fit is unstable and only starts to stabilize when N » 220 as shown in the lower panels.

Once again, in our co-author meeting to finalize the submission version of the manuscript, we decided that an informative legend in Fig. 1 which shows converge to 6 decimal places when 1408 < N < 2816 points would be sufficient to “justify” in a way our choice of N=2200 points (a “round” 100-fold increase in radial grid resolution) – without overloading the reader with additional detail. We have added a short sentence in Sect. 3.1 on page 10580 line 11:

“The legend of Fig. 1 shows that convergence is achieved to 6 decimal places is achieved when the number of interpolation points N is in the range 1408 $\leq$ N $\leq$ 2816 points – i.e. when there is approximately a 100-fold increase in the radial grid resolution (=2200 points). At this resolution, it was found that the standard error associated with the GMM method described in Sect. 3.3 was stable.”

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

We thank the reviewer also for this important point. The uploaded figures were produced at 600 dpi as .eps files. In the AMTD version, the figures are not full page width and the text is, we agree, illegible. We are of course happy to provide higher quality versions with larger axis label fonts as part of the final publication process if AMT proceeds with publication of the manuscript.

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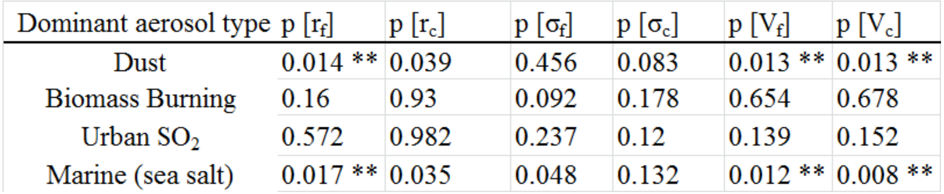

Dominant aerosol type	p [r_f]	p [r_c]	p [σ_f]	p [σ_c]	p [V_f]	p [V_c]
Dust	0.014 **	0.039	0.456	0.083	0.013 **	0.013 **
Biomass Burning	0.16	0.93	0.092	0.178	0.654	0.678
Urban SO_2	0.572	0.982	0.237	0.12	0.139	0.152
Marine (sea salt)	0.017 **	0.035	0.048	0.132	0.012 **	0.008 **

Fig. 1.

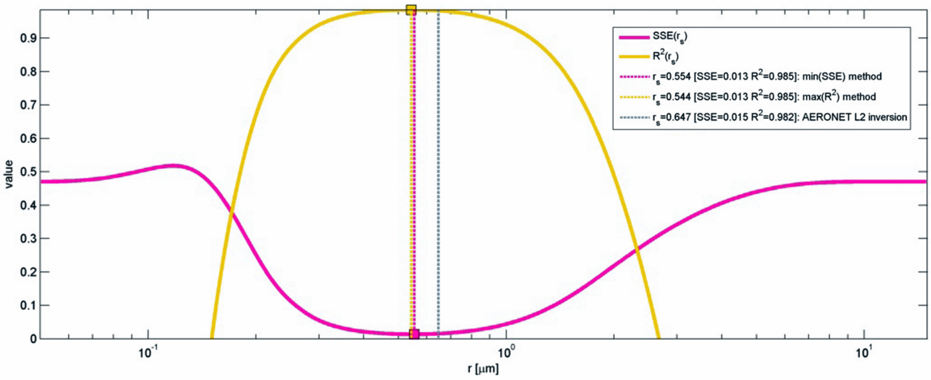

Fig. 2.

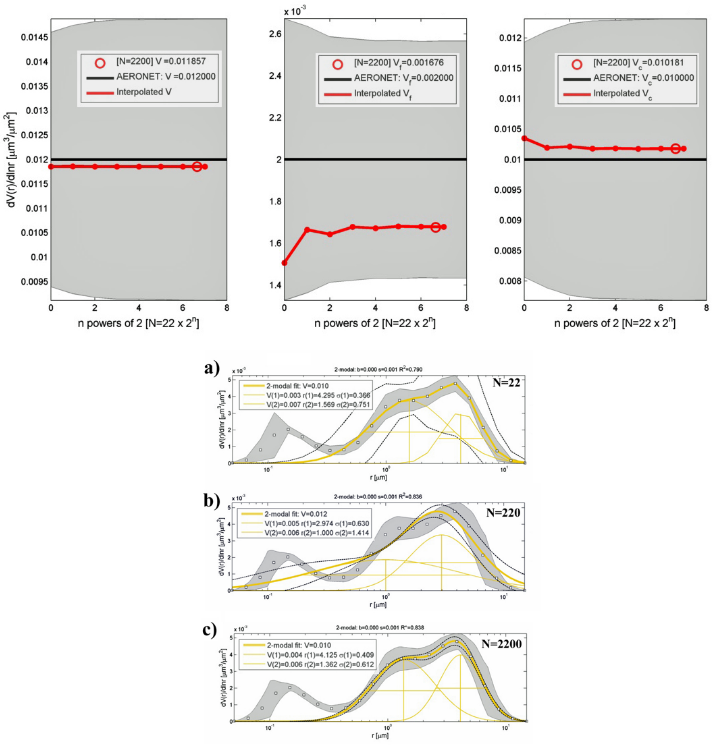
dV(r)/dlnr [μm³/μm²]
[N=2200] V =0.011857
AERONET: V =0.012000
Interpolated V
[N=2200] V_f=0.001676
AERONET: V_f=0.002000
Interpolated V_f
x 10⁻³
[N=2200] V_c=0.010181
AERONET: V_c=0.010000
Interpolated V_c
n powers of 2 [N=22 x 2ⁿ]
n powers of 2 [N=22 x 2ⁿ]
n powers of 2 [N=22 x 2ⁿ]
a)
2-modal: b=0.000 s=0.001 R²=0.790
2-modal fit: V=0.010
V(1)=0.003 r(1)=4.295 σ(1)=0.366
V(2)=0.007 r(2)=1.569 σ(2)=0.751
N=22
b)
2-modal: b=0.000 s=0.001 R²=0.836
2-modal fit: V=0.012
V(1)=0.005 r(1)=2.974 σ(1)=0.630
V(2)=0.006 r(2)=1.000 σ(2)=1.414
N=220
c)
2-modal: b=0.000 s=0.001 R²=0.838
2-modal fit: V=0.010
V(1)=0.004 r(1)=4.125 σ(1)=0.409
V(2)=0.006 r(2)=1.362 σ(2)=0.612
N=2200
dV(r)/dlnr [μm³/μm²]
r [μm]

Fig. 3.