

Anonymous Referee #3

The idea of extending the available set of sun photometer data by data from the GAW filter radiometers certainly is of value. Still I miss a more in-depth rationale why using additional data would be useful. Are these stations closing gaps in the dense network of AERONET stations? Do these filter radiometers provide data that are not available from AERONET? Do these radiometers from GAW in any other way supplement the AERONET data?

Here is some more rationale for explaining the use of the GAW data and the comparison with AERONET, that have been also added in the new text.

The GAWPFR network provides quality assured, high quality spectral AOD data with nearly continuous temporal coverage. Due to its excellent stability (degradation significantly less than 1% /year), recalibrations are done at bi-annual or even less frequencies to minimize data gaps. In contrast to AERONET where station gaps due to recalibrations can be 6 months every 18 months (66% data availability or less), the comparable value for GAWPFR stations is of the order of 90% or more. The main objective of the GAWPFR network is to provide homogeneous data on decadal time scales with the highest possible accuracy. It is installed at global atmospheric watch (GAW) stations to monitor the background aerosol concentrations and thus not to be affected by local changes (urbanisation, etc). Finally, The traceability of GAWPFR is directly to the World Optical Depth Research and Calibration Centre, which was selected by the WMO to act as World reference center for AOD measurements, to which all other networks should be traceable to, including AERONET.

So in a way they are supplementary to the AERONET including some more strict installation criteria according to the WMO and GAW station characteristics.

The fact the LE method is applied to the GAW-PFR data is an insufficient justification to publish this paper, as the LE method has already been presented before, and the authors do not go into any depth to explain the contradictions they find in the results presented in their study. For the most part their text reads more like a summary. I miss the critical scientific evaluation of their findings and how they intend to resolve the contradictions they identified in their study.

Our paper aims to demonstrate the feasibility of applying the LE on GAW-PFR data, so as to extend the product suite of this network. We believe that this first attempt deserves publication since it demonstrates the potential of using LE to expand the capabilities of GAW-PFR network. The new products provided by the LE can be used on satellite validation studies over rural areas that are covered by the GAW-PFR rather than (mostly) urban AERONET stations. For example, clean continental aerosol types are not well represented in AERONET and the GAW-PFR network can add to that. In our opinion, the significance of the LE method application on PFR is high and the study is considered as a first step to this direction.

Even if we agree with the reviewer that a more critical scientific evaluation of the findings is needed, we think that the discussion should include a criticism also on AERONET inversions. These retrievals have not been thorough evaluated/validated in the past. However, AERONET is the reference for all satellite validation studies presented in a vast number of papers that can be found in the literature. We think that we kept a line between GAW-PFR and AERONET

comparison that is enough to demonstrate the potential of the LE on sunphotometry. Moreover, we believe that it is critical to expand the products of other sunphotometric networks that can contribute to satellite validation activities as an independent source of information.

To strengthen the scientific evaluation of our results, we have extended sections 2.2 and 3.3. Discussion on the origin of retrieval's uncertainties has been added together with considerations on how to decrease these uncertainties.

Added text:

As was mentioned, the uncertainties of inversion obtained for $\epsilon=0$ include the errors arising from existence of the null-space and incorrect choice of the refractive index. To estimate influence of this second factor we performed retrievals assuming $mR=1.35$ and 1.55 , while the model value was $mR=1.45$. For Type II aerosol the variation of retrieved parameters was below 10%. Small particles (Type I) are more sensitive to the choice of mR and corresponding variations are up to $\sim 30\%$ for effective radius and up to 15% for volume. However sensitivity of data to the real part of refractive index allows estimation of mR from the measurements, so finally retrieval uncertainty is below 20% for radius and below 10% for the volume, as it follows from Table 2. Influence of the imaginary part was even less significant: choice of $mI=0.01$ instead 0.005 didn't increase errors of retrieval for more than 5%.

The values presented in Table 2 represent the absolute errors of retrieval, while relative changes of parameters can be obtained with lower uncertainty. For example, if the particle effective radius and refractive index don't vary much the uncertainties due null-space and the choice of inversion interval are also quite stable, so relative change of the particle volume can be evaluated with significantly lower uncertainty than given in Table 2.

The numerical simulations in our paper were performed just for two extreme cases, when either fine or coarse mode dominates PSDs to illustrate the main tendency: increase of retrieval uncertainty when the coarse mode becomes predominant. For more realistic scenario wider range of the mode parameters together with refractive index variation should be considered. Such simulations are in progress, but several notions should be made:

- As the problem is underdetermined, the uncertainties can be decreased by narrowing the "search space" for the inversion interval. The spectral dependence of Angstrom parameter allows to separate the contributions of the fine and coarse modes to AOD (O'Neill et al., 2003) and this information can be used to constrain r_{\min} and r_{\max} ranges in the retrieval. Including the spectral dependence of Angstrom in the analysis should improve the retrieval and it will provide the basis for characterization of retrieval uncertainties.

- Considering AERONET retrievals as a "true" and accumulating sufficient statistics for different types of aerosol it is possible to introduce correction to the LE retrieval to decrease uncertainty, especially for big particles.

Both of these studies are in a progress and the results will be presented as a separate publication.

The authors provide a comparison of results of the LE method applied to filter radiometer data and AERONET CIMEL data. Even though the same algorithm is used, there are differences, and I do not understand the reasons that cause these differences.

Four AODs from PFR and seven AODs from Cimel have been used, so the CIMEL retrieval should be more accurate. Besides, as Table 2 demonstrates even 5% difference in AOD will provide a discrepancy in the retrievals, so difference in error distribution through channels in both instruments may also be the reason. Corresponding comment is added to the manuscript.

Could one reason be the fact that the authors only use a few measurement cases for their study? I am missing a more detailed discussion of possible reasons. The authors provide an error analysis on the basis

of synthetic data. The range of input parameters (two size distributions only?) is clearly insufficient for a robust, statistically significant error analysis. The error analysis presented in section 3.3 reads as if it was only added because there was no choice to avoid the topic of error analysis. The section is short, does not cover the real problems one is facing in error analysis, does not treat a sufficiently large number of aerosol scenarios that would allow the reader judge the quality of the results of the experimental data. Using just one refractive index in the error analysis is clearly insufficient, particularly in view of the complex aerosol cases presented in this paper. Figure 6 clearly shows the retrieval errors that occur at high solar zenith angles. I may have missed or not fully understood that section of the paper in which they authors point out to this effect and discuss in how far their evaluation of the use fullness of the LE method is biased by the fact that retrieval results need very careful correction of data taken at high solar-zenith angles. Why do the results for AERONET- LE differ so much to the PFR-LE results? It is the same method and the measurement time was nearly the same. Is it because different measurement wavelengths were used?

We hope that the two previous replies to the comments are covering this issue. The section 3.3 is extended and discussion of the retrieval errors is added. For direct comparisons of AOD retrievals by the two instruments we believe that the measurements period was enough for assessing the differences. In addition, we provide a link among the two networks for AOD retrievals. Comparing the use of the LE method with both CIMEL and PFR we end up in differences that can be explained by the fact that LE method with the CIMEL in principle should be more accurate due to the use of the seven instead of four wavelengths. Yes we agree that a period including more simple (or a variety of) aerosol cases would provide the opportunity to assess the differences for different aerosol cases. What it is planned for the future is the use of the LE method in various CIMEL instruments around the globe with different aerosol characteristics.

We agree that the use of one refractive index on the analysis was insufficient, so the new text in section 3.3 (provided above) includes a discussion and a sensitivity analysis on this issue.

I am also puzzled by the underestimation of the volume concentration from the LE method compared to the volume concentration that is obtained with the standard AERONET algorithm, see figure 8. Again: I may have missed the main point in the paper that explains it. Could the difference be because the AERONET standard algorithm provides particle size distributions that separate fine and coarse mode fractions?

This is due to the fact that the standard AERONET algorithm uses angle scanning of sky radiance, while LE uses only AODs measurements. The AOD measurement kernels are less sensitive to the coarse mode, so this is the reason that the volume of big particles is underestimated. Text was also included in the new document.

Does the LE algorithm do the same or does it only provide an "average" size distribution, i.e. it does not separate into fine and coarse mode? In that case I would almost naturally expect differences as the averaging (mean particle size distributions without distinguishing between coarse and fine mode) may create a bias.

In principle, LE provides the particle size distribution as intermediate result, so the contributions of the fine and coarse modes can be separated. However, the uncertainties that are introduced using such separation are high and we don't present corresponding results at the moment. Instead we are

preparing the correction of the coarse mode retrieval, considering AERONET retrieval as “true”. We plan to present the results soon as a separate publication.

Given the fact that there are systematic difference between results from the LE method and the standard AERONET retrievals I am missing a more detailed description of the filter radiometer technical features. Which wavelengths are used, what is the field of view of the instruments, which instruments parameters could be reasonable for the differences?

We think that the reason for the difference between LE and standard AERONET retrieval is quite fundamental. AERONET uses angle scanning of sky radiance, so particle sizes of radii up to 15 μm can be retrieved, while LE uses only AOD measurements and the retrieval ability of the particle sizes is constrained to smaller radii, since the corresponding kernels lose their sensitivity to particles with radii above 5 μm .

The results of figure 10 concern me: minimum errors for the coarse mode size distribute are at least 30 - 50%. Could these errors be the result of the limited “inversion range” which seems to be lower than the 15 micrometer that is usually used by AERONET standard retrieval? Please comment on this and provide an explanation.

This is the property of the kernels used. Without sky radiance angle spectrum the retrieval is insensitive to significant part of the coarse mode. As we have mentioned, we are preparing a correction procedure to decrease these errors.

Figure 11 is not needed. It would be more useful to show a map of the Athens area in which the location of the instruments is shown. This might help to understand a bit better why there are differences of the results from the two methods (LE and AERONET algorithm).

Detailed comments:

I am missing a summary of the specific features of the LE method, how it compares to the standard AERONET algorithm and what work has been done before. It is hard to judge how many “new findings” are presented in this paper, except for the fact that the data from the GAW radiometers can be included into existing AERONET data.

Description of LE approach is extended, still we don’t want to do it very detailed, because it will repeat our previous publication.

Fig 1: please merge figures 1a and 1b into one figure. This would make it much easier to see the correlation between changes of AOT and changes of Angstrom exponents.

Please also show the AERONET and Angstrom exponents from AERONET in a separate plot.

I find figure 2 confusing, it does not tell me a lot expect that there was a strong pollution event around 18/19 July. The logarithmic scale for radius does not really help to understand the time series.

Figure 4, a: is it an instrument problem that causes the significant differences of AOT measured with CIMEL and the PFR at low AOD at 500 nm? It does not become clear in the text in how far this bias causes the problems for effective radius and volume concentration.

From the analysis of the influence of the random errors in the input data presented in section 3.3 we can expect that a 5% difference in optical data may lead to 10-20% difference in retrieved parameters. The text was included in the new document.

Figure 5: please show several representative error bars for the PFR-LE, AERONET-LE and Angstrom.

In section 3.3 we discuss the uncertainty of the retrieval. To specify the uncertainty for every point we need to simultaneously analyze the spectrum of the Angstrom exponent, because the absolute uncertainty depends on the contribution of the coarse mode. Such option is not ready yet, so we can expect only the worst case and consider the uncertainty of the volume and effective radius retrieval as high as ~60%. But as discussed in section 3.3 the errors of relative changes of the parameters are actually significantly smaller.

The reference list needs considerable extension with references that deal with AERONET measurements, quality assurance tests, validation studies. There have been no comparisons of AERONET results to airborne in-situ measurements or studies that compare lidar retrievals to AERONET data? AERONET presents the current standard in terms of aerosol data products, so there should be at least a summary of the current studies regarding quality assurance of AERONET data. To my knowledge there are not many studies that compare AERONET results to results from other instruments, so the literature search should be quite straightforward. The results from this search would be the starting point to judge the quality of the GAW-PFR results. Publications that show the current status of the GAW filter radiometers in terms of previous measurements and main outcomes should be presented as well. At the moment most of the literature is quite general and does not help understanding the main results of this study.

The reference list have been expanded. A summary of the AERONET measurement status concerning the quality assurance, other validation studies and finally a review of the GAW-PFR network have been included in the new manuscript.

The following comments have been taken into account.

The reference Hansen and Travis 1974 is very old. Are the findings presented there still valid in view of the enormous progress that has been in the past 30 years of aerosol research with radiometers?

Page 104, line 21: course » coarse

Page 104, line 26: excludedfrom » excluded from

Page 105, line 1: utily???

Page 105, line 21: quantify the "residual"

Page 108, line 17: resultsfrom » results from