Anonymous Referee #1

Received and published: 18 February 2014

The paper is on the retrieval of aerosol properties: from spectral AODs the effective radius (reff) and the volume concentration (Vc) are derived. The advantage of the LE- approach (linear estimation) is that it is based on spectral AOD measurements only, i.e., no principal plane and/or almucantar measurements are required. The authors applied the LE-method to coincident and co-located measurements of the PFR- and the Cimel-radiometers. The results are inter-compared, and compared to results of the routine AERONET inversion scheme. The differences between the three types of evaluation are discussed with respect to the solar zenith angle and the AOD. The agreement varies between good and acceptable depending on the aerosol-parameter.

The paper is clearly structured and well written. However, the scientific reasoning must be extended and must be more convincing. A few suggestions of desirable/mandatory amendments follow. If the missing arguments are provided, the paper can be published.

Comments

The scientific concept of the LE-retrieval obviously is, that AOD is somehow related to Vc (this is plausible, as a higher AOD normally is associated with larger Vc) and that the spectral dependence of the AOD is somehow related to reff (this is also plausible, the Angström exponent is well known). So, in other words, the products of the retrieval are the same as conventional products (AOD and Angström), the only difference is that this information is expressed by two other variables. As a conclusion it is mandatory to explain the advantages of the "reff + Vc "-concept over the "AOD + Angström" approach.

Actually, the relationship between AODs and volume is more complicated, because the Particle Size Distribution shape can vary a lot for given AOD, although it depends strongly on the AOD spectral behavior (Ångström). In our approach we take advantage of this dependence and represent the particle volume as a linear combination of all AODs (up to seven for AERONET data). We do the same for the effective radius, since it also depends on the AOD spectral behavior/Ångström. So, the inversion of AODs to particle properties using the LE technique provides more information than just the initial AODs + Ångström. (Moreover, the LE technique allows to calculate other parameters such as number, surface particle density and also AOD at other wavelengths.)

a. Modelers certainly would prefer reff and Vc, but if the errors are too large, the supposedly benefit disappears. In this context, the error analysis in Sect. 3.3 is quite useful, but should be extended (the variability of aerosol distributions is large! and includes more parameters than the authors have varied).

b. Is an error of 60% acceptable for modelers?

c. Can the operator identify whether a case I (small, acceptable errors) or case II (large, unacceptable errors) situation had occurred?

a. The detailed analysis of the errors will be presented in our coming manuscript, where we introduce the technique for correction of uncertainties, assuming AERONET retrievals as "true". That way the operation of inversion algorithm is significantly improved, especially for big particles. So error analysis will be given for that upgraded algorithm. In this study we intended to demonstrate the feasibility of particle parameters estimation from four measured AODs, and to illustrate the main tendency - increase of uncertainty for the coarse mode predominance.

Sections 2.2 and 3.3 have been extended and the paragraph above have been added to the new text. Discussions the origin of retrieval uncertainties are added together with consideration the ways to decrease these uncertainties.

b. From our discussions with modelers we conclude that even 60% uncertainty can be acceptable in many cases. We should also recall that:
- The relative change of parameters can be obtained with significantly smaller uncertainties than absolute one.
- The errors of retrieval can be partly corrected, considering AERONET retrievals as "true". We are working on this approach at a moment.

Corresponding comment is added to the text

c.Yes, such information is available from Angstrom exponent measurements. From long-term statistics of AERONET measurements we conclude that for $AE(440\text{-}870)>1.5$ more than 70% of AOD at 500 nm is attributed to the fine mode, so it can be taken as a case I. Similar for $AE<0.5$ the coarse mode contributes above 70% to AOD, so it can be taken as case II. For $0.5<AE<1.5$ the errors are intermediate. We are working on more solid base of errors characterizing using the spectrum of LE.

The paper's message is that a "simple approach" can provide useful aerosol information. Whether the retrievals are correct or not and which one is the best, is not answered as no independent data are available (i.e., no validation is possible), the paper only points out, how large the differences between the three retrievals are. Additional to the assessment of the accuracy of the microphysical parameters (as mentioned above), probably a review of previous papers will help. I assume that there are validation experiments for the routine AERONET-retrieval available. Then, comparisons with these results can serve as a benchmark. As more optical data are included in the AERONET-retrieval, it can be expected that these results are in principle more accurate.

The LE procedure is applied to pure spectral particle extinction data, no sky radiance information (phase function) is available in contrast to the AERONET inversion method. So, the AERONET inversion retrieval products clearly represent the quality standard. An assessment of AERONET product accuracy is provided in Dubovik et al. (2000), where the inversion is tested on synthetic data considering different aerosol types (water-soluble aerosol, biomass burning and dust) and taking into account measurement random error and systematic errors due to miscalibration, azimuth angle pointing and surface reflectance. Therein, the accuracy of the retrieved volume size distribution ($dV/dlnr$) for dust is estimated to 35% for $0.1\mu m<r<7\mu m$ and 35-100% for $r<0.1\mu m$ and $r>7\mu m$. Note that this analysis was done before the non-spherical particles modelling was included in the AERONET inversion scheme. Dubovik et al. (2006) refer that the new inversion scheme, taking into account the non-spherical shape of the particles, provides better fittings of the sky radiances and AODs used for the retrieval for the case of dust aerosol, to the level of the measurement accuracy, whereas the spherical model results in at least double fitting errors. Validation studies of the AERONET volume size distribution for dust aerosol using independent data show varying agreements. For example, in Toledano et al. (2011) the comparison with in-situ airborne data during the SAMUM-II campaign, showed good agreement for the shape of the size distribution, although the effective radius of the coarse mode from AERONET was smaller than the one measured with the in-situ instruments on-board. Muller et al. (2010) report more significant differences for both fine and coarse mode of dust particles compared to aircraft measurements and ground-based in-situ measurements during SAMUM-I.

1. Dubovik, O., A. Smirnov, B. N. Holben, M. D. King, Y. J. Kaufman, T. F. Eck, and I. Slutsker (2000),Accuracy assessments of aerosol optical properties retrieved from Aerosol Robotic Network (AERONET) Sun and sky radiance measurements, J. Geophys. Res., 105(D8), 9791–9806, doi:10.1029/2000JD900040.

2. Dubovik, O., et al. (2006), Application of spheroid models to account for aerosol particle nonsphericity in remote sensing of desert dust, J. Geophys. Res., 111, D11208, doi:10.1029/2005JD006619.

3. Toledano, C., et al. (2011), Optical properties of aerosol mixtures derived from sun sky radiometry during SAMUM 2, Tellus, Ser. B, 63, 635–648, doi:10.1111/j.1600-0889.2011.00573.x.

4. Mineral dust observed with AERONET Sun photometer, Raman lidar, and in situ instruments during SAMUM 2006: Shape-independent particle properties. D. Muller, B. Weinzierl, A. Petzold, K. Kandler, A. Ansmann, T. Muller,2 Tesche, V. Freudenthaler, M. Esselborn, B. Heese, D. Althausen,2 Schladitz, S. Otto, and P. Knippertz

It is shown, that the agreement of the AODs of the two radiometers is excellent. As a consequence, it is expected that the retrieval of r eff and Vc gives similar results: "same input" plus "same retrieval" should provide the "same result". From this point of view the differences between the two LE-results are surprising, and only the differences to the AERONET-inversion are relevant. Therefore, the authors must give more details of the data, e.g., it is not clear how many spectral AODs (at what wavelengths) are involved in the PFR-LE and the Cimel-LE. This might explain the differences. Another reason can be the large sensitivity to even small measurement errors as briefly mentioned in Section 4. Anyway, this issue must be clarified.

Yes we agree with the reviewer that in a way it is surprising. Main reason for this difference is the use of just four AODs from PFR and seven AODs from Cimel, so retrieval from Cimel data should be more accurate. Besides, as Table 2 demonstrates even 5% difference in AOD will provide discrepancy in retrievals, so difference in error distribution through channels in both instruments may also be the reason. The discussion will be inserted in the new text.

The measurement period was dominated by a complex meteorological situation when two quite different aerosol types were mixed. As a consequence, it is certainly difficult to assume a reasonable refractive index for the atmospheric column. Though the authors state that the relationship between AOD and microphysics is not very sensitive to the refractive index, this issue should be discussed in some detail. So not only the dependence on the solar zenith angle and the AOD should be highlighted but also the dependence on the "degree of mixing" (even during the dust event it was not pure dust?).

We have added corresponding comment in section 3.3

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

By the way: I do not understand the sentence "despite the presence of a strong dust event" in the abstract: why is the accuracy of the AOD influenced by the dust event?

It was written that way mainly because dust particles are characterized by larger effective radius and it leads to enhanced uncertainty of the retrieval. It was clarified in the abstract.

The retrieved r eff is always somewhere between the two modes shown in Fig. 2. This is expectable, but the question comes up how valuable this information is R eff neither describes the coarse mode nor the fine mode, and the uncertainty of r eff might be large (see above)

This is a very important question. From our discussions with modelers we conclude that even such integral effective radius is important in many situations. At the moment we are trying to expand the method in order to implement the possibility to estimate particle parameters separately for fine and coarse mode. However this is quite challenging, because in the case of the PFR only 4 input data are available. Application of the method to other direct sun spectroradiometers or sun-photometers able to measure AOD in more wavelengths could probably provide improved information.

It would be nice to have another measurement example when only one aerosol type was present. I don't know for which period coincident measurements are available. Maybe no further data are available.

In the current manuscript we just aimed to show the feasibility of LE code application to retrieve Reff and Vc with an uncertainty estimation taking into account a coordinated comparison of the two instruments (only for that short period of time). At the moment another manuscript is in preparation, where we consider a large number (~75000) comparisons between the operational AERONET and LE codes for different aerosol types (and locations) to get sufficient statistics.

The number of references should be increased: a few examples of sun photometer measurements (of dust events) and their inversion should be cited and commented

We have adde the following text and references:
AERONET products for dust aerosol worldwide are presented (along with other aerosol types) in AERONET climatology of Dubovik et al., (2002). Furthermore, the study of Dubovik et al. (2006) demonstrates the non-spherical model included in the AERONET inversion algorithm that accounts for the non-sphericity of the dust particles. The AERONET dust product is expected to be further enhanced in the new GARRLiC algorithm which combines the sunphotometric measurements with lidar measurements (Lopatin et al., 2013). Validation studies comparing column-averaged volume size distributions obtained from sunphotometer measurements with direct in situ measurements at some locations (aircraft as well as surface-based) (Haywood et al., 2011; Toledano et al., 2011; Gerasopoulos et al., 2007;), are key to helping systematically validate microphysical inversion products [IPCC AR5 (2013), Chapter 7, p603] from AERONET and GAW.

Dubovik, O., Holben, B., Eck, T. F., Smirnov, A., Kaufman, Y. J., King, M. D., Tanré, D., and Slutsker, I.: Variability of absorption and optical properties of key aerosol types observed in worldwide locations, J. Atmos. Sci. 59, 590–608, 2002.

Dubovik, O., et al. (2006), Application of spheroid models to account for aerosol particle nonsphericity in remote sensing of desert dust, J. Geophys. Res., 111, D11208, doi:10.1029/2005JD006619.

Gerasopoulos, E., Koulouri, E., Kalivitis, N., Kouvarakis, G., Saarikoski, S., Mäkelä, T., ... & Mihalopoulos, N. (2007). Size-segregated mass distributions of aerosols over Eastern Mediterranean: seasonal variability and comparison with AERONET columnar size-distributions. Atmospheric Chemistry and Physics, 7(10), 2551-2561.

Haywood, J. M., et al., 2011: Motivation, rationale and key results from the GERBILS Saharan dust measurement campaign. Q. J. R. Meteorol. Soc., 137, 1106–1116.

Lopatin, A., Dubovik, O., Chaikovsky, A., Goloub, P., Lapyonok, T., Tanré, D., and Litvinov, P.: Enhancement of aerosol characterization using synergy of lidar and sun-photometer coincident observations: the GARRLiC algorithm, Atmos. Meas. Tech., 6, 2065-2088, doi:10.5194/amt-6-2065-2013, 2013.

Toledano, C., Wiegner, M., Groß, S., Freudenthaler, V., Gasteiger, J., Muller, D., Muller, T., Schladitz, A., Weinzierl, B., Torres, B. and O'Neill, N. T. (2011), Optical properties of aerosol mixtures derived from sun-sky radiometry during SAMUM-2. Tellus B, 63: 635–648. doi: 10.1111/j.1600-0889.2011.00573.x

A few further comments in brief (partly already implicitly mentioned above): The basic idea (in terms of radiative transfer) of the LE should be briefly reviewed, summarized and compared to the fundamentals of the routine inversion scheme. This is essential to understand the limitations and the potential of the LE.

We extended 2.2 section with the description of algorithm. Still not fully repeating the detailed description given in our previous publication.

The instruments should be described in more detail in particular the spectral channels (see comment above). The comments in the introduction are not sufficient and in Section 2 the relevant information is also missing.

We have included more information on the instrument describing section as the reviewer suggests.

	CIMEL	PFR
Central Wavelengths	1639.8, 1017.7, 870, 672.7, 500.1, 438.5, 379.7 and 339 nm	861.6, 500.5, 411.4 and 367.6 nm
FWHM	2nm for 340nm 4nm for 380nm 10nm for the other wavelengths	5.4, 5.0, 4.5 and 3.8 nm
Field Of view	1.2°	2.5°

Page 106, line 1: why is the upper limit of the inversion windows 10 μ m and not 15 μm as in case of Cimel?

Kernel functions for AODs used are not sensitive to radii above 10 μm and increase of upper limit up to 15 μm only spoils the retrieval. In the Cimel retrieval as the angular scanning of sky radiance is used, the consideration of larger radii is possible. The explanation was also included in the document.

Page 113: the equation of log-normal size distribution may be given, even it is well known.
But actually eq.5 contains expression for log-normal size distribution. We think it is sufficient for reader.

I would rather appreciate more equations describing the LE-formalism.

We added some additional description, but adding formalism would just repeat our previous publication, which we tried not to do.

All the following remarks have been taken into account and commented or changed in the new manuscript

Most figures must be optimized: larger labels, check description of colors in Fig. 6, Fig. 11 has the wrong number, "log" should be "ln" in Fig. 2, and the y-axis scale should be changed in Fig. 6 (starting at 0.15 or so).

Page 109, line 12: "The best-fit line" This sentence should be re-phrased; it is not clear, why the LE is mentioned here.

The abstract can be shortened; don't give too many details.

Page 102, line 17: it's an ill-posed problem for Cimel as well.

Page 108, line 15: don't describe wavelength-differences in percent; it does not make sense.

Finally: the conclusions shall be reviewed: certainly a few duplications with the abstract can be avoided