

Dear Referee #2

many thanks for your comments which have allowed to improve the paper as demonstrated in the revised manuscript. The lack of a rigorous error analysis (uncertainty discussion) represents a weak point of the paper, according to your comments. Systematic uncertainties from different sources have been taken into account in the revised manuscript. More specifically, we have allowed to the input parameters used to invert lidar profiles to vary in prescribed ranges, to explicitly calculate the effect of their variations on Angström exponents and differences. The iterative numerical procedure suggested by Di Girolamo et al., (1994) and Marengo et al. (1997) was adopted in the revised manuscript to invert the lidar signal profiles under the constraint of a measured total AOT. The used methodology and the uncertainty discussion have been presented in Section 2.2 of the revised manuscript. An excerpt of Section 2.2 of the revised paper is reported below:

“ 2.2 Aerosol parameters from lidar measurements

The UNILE lidar system was designed to derive vertical profiles of the aerosol extinction ($\alpha(z)$) or backscattering ($\beta(z)$) coefficient at 355 nm, 532 nm and 1064 nm, respectively and of the volume depolarization ratio ($\delta(z)$) at 355 nm during day time measurements. The approach proposed by Fernald (1984) and Klett (1985), which requires an *a priori* value of the aerosol extinction-to-backscatter ratio (also referred to as the aerosol Lidar Ratio, LR), is commonly used to invert lidar signal profiles and extract aerosol extinction and backscattering coefficient profiles. The assumption that LR is known *a priori* is likely the largest source of systematic error within this lidar inversion procedure. However, this uncertainty can be largely reduced if additional information is available. Takamura et al. (1994) considered the possibility of removing the indeterminacy in LR by combining lidar data with independent measurements of the aerosol optical thickness. Then, Di Girolamo et al. (1994) and Marengo et al. (1997) suggested an alternative inversion technique, which through an iterative procedure allows one to determine $\alpha(z)$ and $\beta(z)$ by using as boundary conditions (1) the AOT of a selected altitude range and (2), as in the Fernald-Klett approach, the total backscattering coefficient β_T (due to molecules (β_M) and aerosol (β)) at a far-end reference height z_f . This last approach was used in this study to extract aerosol extinction profiles at 355 nm, 532 nm, and 1064 nm, respectively from UNILE lidar measurements. AOT values at the lidar wavelengths were retrieved from AERONET sun/sky photometer measurements co-located in space and time. An AERONET sun/sky photometer operates at the UNILE lidar site since the year 2003 and it provides AOTs with accuracy of ± 0.01 , according to Dubovik et al. (2002). Hence, it was required that the AOTs calculated from the aerosol extinction profiles should not exceed (within ± 0.01) the corresponding AOT values retrieved from co-located sun/sky photometer measurements. More specifically, the lidar AOTs at 355 nm, 532 nm, and 1064 nm, respectively, were calculated from the corresponding $\alpha(z)$ profiles by assuming that $\alpha(z)$ values did not vary with altitude below the height (z_i) where the lidar system was estimated to achieve full overlap. The full overlap height varies within 0.5-1.0 km a.g.l. for the lidar system of this study. Note that the planetary boundary layer (PBL) height varies within 0.4-1.0 km a.g.l. at the monitoring site of this study (De Tomasi and Perrone, 2006; De Tomasi et al., 2011). Aerosol particles are well mixed within the PBL and as a consequence, it is reasonable to assume that $\alpha(z)$ values did not vary with altitude below ~ 1 km a.g.l.. The far-end reference height was chosen, for each profile, in a region where the lidar signal followed the molecular profile and hence, it was assumed $\beta_T(z_f) \equiv \beta_M(z_f)$. Note that the assumption of an altitude independent lidar ratio to retrieve $\alpha(z)$ profiles was still necessary for the iterative procedure used in this study. A discussion on this assumption is reported in Sect. “Sensitivity test on the lidar ratio vertical profiles for 28 July 2011”. Uncertainties in the retrieved $\alpha(z)$ profiles include statistical uncertainties due to the presence of noise on the received lidar signals and systematic uncertainties as the ones due to the assumed molecular profile, the reference backscattering coefficient value, the total measured AOT, the AOT contribution of the atmospheric

layer below the overlap height z_i , and the lidar ratio variability. Radiosonde measurements at the meteorological station of Brindisi (<http://esrl.noaa.gov/raobs/>) that is 40 km north-west of the monitoring site of this study were used to define air density vertical profiles and decrease the uncertainty associated to the Rayleigh scattering calibration. The uncertainty on the reference backscattering coefficient value was accounted for by assuming that the aerosol backscattering coefficient varied from a nil value up to $5 \times 10^{-7} \text{ (m sr)}^{-1}$ at $z = z_f$. The error on the AOT contribution of the lowermost atmospheric layer located at $z < z_i$ a.g.l. was accounted for by allowing to the AOT contribution within the $(z_i - z_f)$ atmospheric layer (AOT_1) to decrease up to 20% of a reference value $\text{AOT}_{1,\text{ref}}$. To this end, a 2-step numerical procedure was used. In the first step, extinction coefficient profiles at the lidar wavelengths were calculated from the inversion of the lidar signals through the implemented iterative procedure, by setting that extinction coefficients did not vary with altitude from the ground up to the overlap height z_i . Then, the AOT contribution of the $(z_i - z_f)$ atmospheric layer ($\text{AOT}_{1,\text{ref}}$) at each lidar wavelength was calculated. In the second step, extinction coefficient profiles were calculated from the implemented iterative procedure by allowing to AOT_1 to decrease up to 20% of the determined reference value $\text{AOT}_{1,\text{ref}}$. In fact, the condition that $\alpha(z)$ values did not vary with altitude from the ground up to z_i could be responsible for an underestimation of the AOT contribution of the lowermost atmospheric layer (AOT_2). The inversion of the lidar signals through the implemented iterative procedure is not demanding much computation time so that a few thousand extinction profiles at 355 nm, 532 nm, and 1064 nm, respectively were easily generated by changing boundary conditions. Angström exponent profiles were calculated from the generated extinction profiles. The mean extinction profile at each lidar wavelength was then calculated by averaging all corresponding extinction profiles determined by the iterative procedure and the $\alpha(z)$ uncertainty was set equal to one standard deviation of the mean value. A similar procedure was used to calculate mean profiles of Angström exponents and corresponding uncertainties. Angström exponent differences were calculated from Angström exponent mean profiles. Corresponding uncertainties were calculated by the law of error propagation”

Details

Page 4, lines 1-10: What about the other combined lidar/photometer methods (GARRLIC, Lopatin et al., AMT 2013, POLIPHON, Ansmann et al., ACP 2012)?

The GARRLIC and POLIPHON methods have been mentioned in the revised manuscript and corresponding references have been added.

Page 4, line 27: There are a variety of O’Neill papers (beginning in Appl. Opt. 2003 or even earlier) dealing just with the slope of the AOT to get the fine-mode and coarse mode contributions to total AOT. One of these methods is now a standard retrieval in AERONET. Any comment to that?

Both the paper by Schuster et al. (2006) and the one by Gobbi et al. (2007), which were on the manuscript reference list, are based on the papers by O’Neill, as it was clearly stated in both papers, where an overview of the O’Neill work is also reported. As a consequence, we thought that it was not necessary to mention the O’Neill works in the manuscript. However, the paper by O’Neill et al. (2003) has been added to the references of the revised manuscript.

Page 7, line 12: Please improve: aerosol extinction-to-backscatter ratio, not aerosol to-backscatter ratio.

Done

Page 7, line 18-27: Please specify (quantify) the uncertainty in the backscatter retrieval

at 1064 nm.

See below.

.....Estimate by varying the assumption on the AOT contribution of the lowermost atmospheric layer (below the overlap height), i.e., for the layer of 1 km which is not covered by lidar,.....

The methodology used to vary the assumption on the AOT contribution of the lowermost atmospheric layer has been discussed in Section 2.2 of the revised manuscript (see above).

.....and please quantify then also the error in the lidar ratio estimates...

The used iterative procedure to invert the lidar equation provides the lidar ratio values that have been determined to fulfil boundary conditions. Mean lidar ratio values and corresponding standard deviations will be reported in Table 2 of the revised manuscript.

Page 7, line 18-27: Please specify (quantify) the uncertainty in the backscatter retrieval at 1064 nm.

.....Furthermore, at 1064 nm the backscatter coefficient solution is very insensitive to the lidar ratio input, but very sensitive to the reference value. This means that the uncertainty in the 1064 nm lidar ratio can be very large, when the AOT of the AERONET Cimel is matched within a given AOT range (± 0.1). I speculate the error is already in the range of 100% for the column lidar ratio caused by uncertainty in the 0-1km AOT contribution. And when one varies the reference value (calibration value in the free troposphere), too, the overall uncertainty will be higher by another factor of 2, I could imagine. What reference value did you use for the different wavelengths? Please state!. At 1064nm this reference value should then be varied by plus/minus a factor of 10 in the uncertainty analysis. In this way, the uncertainty in the backscatter coefficient profile and in the column lidar ratio can be easily quantified, as well as the consequences (uncertainties) for the Angstrom exponents.

We believe that the discussion in Section 2.2 of the revised manuscript (see above) answers all your questions. However, we report in the following the methodology we have commonly used when the reference height uncertainty was quite large at one lidar wavelength. As mentioned, the calibration height must fulfil the criterion that at this altitude the aerosol backscattering coefficient is negligible compared to the molecular backscattering coefficient value. The problem concerning the z_f selection arises in the case of low signal-to-noise ratio (SNR) at the calibration height. For this situation, the relative error of z_f may be quite large. Note that in multi wavelength lidar systems, the SNR at the calibration height depends on the wavelength of the lidar signal and as a consequence, the relative error of the calibration height may be large at some wavelengths and of the order of few percent at the others. More specifically, in Nd-YAG based lidar systems, as the one used in this study, the SNR at the calibration height is on average larger at 355 and 532 nm than at 1064 nm. When we detected a low SNR at the calibration height z_f of the 1064 nm lidar signal, the procedure reported below was used to decrease the calibration height error. In the first step, the 1064 nm extinction coefficient profile at a reference height (z_f^*) where the aerosol contribution was not negligible was estimated from the corresponding 532 nm extinction coefficient by assuming a height-independent Angström exponent (α). In particular, the α value was assumed to be equal to the 500/1020 nm Angström exponent retrieved from AERONET sunphotometer measurements co-located in space and time. Then, once the 1064 nm- $\alpha(z_f^*)$ value was known, the corresponding

aerosol extinction profile was re-calculated from the lidar equation (Tesche et al., 2008; Navas-Guzman et al., 2011).

Page 11, line 6: I trust the lidar ratio at 355 nm (80sr), and also at 532nm (70sr), but I do not trust the 1064nm lidar ratio. What is the uncertainty here (see discussion above)?

Lidar ratio values and corresponding uncertainties will be reported in the revised manuscript.

Page 11, line 20 to page 12, line 16: All statements are speculative without uncertainty numbers, for the Angstrom exponent and the Delta Angstrom value. Please provide uncertainty numbers and then a save, tentative argumentation, avoid speculation.

As mentioned, an improved evaluation of uncertainties will be provided in the revised manuscript.

..... Volume depolarization ratios are at all 1% or lower. Such low volume depolarization ratio (even at 355nm) do not indicate the presence of coarse dust particles.

Particle depolarization ratios will be shown in the revised manuscript. However, depolarization ratios depend on the percentage contribution of coarse dust particles and on the optical property changes that have undergone from the source area up to the monitoring site.

.....and the trajectories

do not support any significant transport of Saharan dust to Lecce. So all this is speculative, the scientific value of the entire discussion here is close to zero.

The following comments have been added in the revised manuscript :

“ Figure 4a shows the pathways estimated at 13:00 UTC of the ten day backtrajectories with arrival heights at 1, 2, and 4 km a.g.l.. The time evolution of the altitude of each backtrajectory is plotted in Fig. 4b. Northern America was the source region of the air masses arriving from 1 up to 4 km a.g.l.. The air masses arriving at 4 km a.g.l have travelled at altitudes varying within 3-6 km a.g.l. before reaching south-eastern Italy. While, the air masses at 2 km arrival height crossed north western Africa before reaching south-eastern Italy and as a consequence, they **have likely picked up lofted desert dust particles. Dust particles lifted up to few kms a.g.l. are commonly subject to long range transport.** To this end, it is worth mentioning that the true colour images from the Moderate Resolution Imaging Spectroradiometer (MODIS) on board the Terra and Aqua satellite, have revealed the presence of Africa dust particles over the south western Mediterranean both at 11:35 UTC (<http://lance-modis.eosdis.nasa.gov/cgi-bin/imagery/single.cgi?granule=T112071035>) and 12:10 UTC (<http://lance-modis.eosdis.nasa.gov/cgi-bin/imagery/single.cgi?granule=A112071210>) of 26 July, 2011. Figure 4b indicates that the 2 km arrival height backtrajectory has likely crossed the south western Mediterranean on 26 July. The BSC-DREAM model also supports the advection of a small amount of Sahara dust particles over south-eastern Italy....”

Page 13, from line 20 downward: again , just speculations without uncertainty values.

Figure 2: The extinction coefficients are large and pronounced in the layer above 3 km, and this layer has sharp lower edges and pronounced variations with height.

Vertical profiles of the potential temperature and relative humidity that were retrieved from radiosonde measurements performed at the meteorological station of Brindisi (<http://esrl.noaa.gov/raobs/>) on 28 July at 11:00 UTC, will be provided in the revised manuscript to

support the vertically inhomogeneous layering of aerosol particles revealed by Fig. 2a and to better understand the changes with the altitude of aerosol properties.

..... Can such structures be preserved after almost 10000 km of air mass transport?

The sentence: “.....were likely due to anthropogenic pollution from Northern America....”

has been replaced in the revised manuscript with the following one:

“...were likely due to anthropogenic pollution lifted up to altitudes $z > 2$ km a.g.l. and then transported over south eastern Italy...”

The Angstrom exponents show a systematic trend (monotonic increase) with height, seems to be a bias. Please provide the respective uncertainty analysis. To be clear, I do not believe in any of the shown results in this figure. The reader needs uncertainty ranges for all parameters so that he/she can draw own conclusions from the presented plots.

Figure 3: The graphical framework is convincing, but the lidar results are not. Uncertainty bars have to be provided in the revised version.

Uncertainty bars were provided in Fig. 2b and 2d and in Fig. 3. However, an improved evaluation of uncertainties will be provided in the revised manuscript.

Page 14, line 16: The use of the altitude independent lidar ratio is just ANOTHER weak point of the analysis, not the only one! The rather uncertain 1064nm backscatter retrieval is the main error source. And as already said, I do not believe that desert dust was dominating the optical properties in the lower troposphere, so lidar ratios around 55sr are not justified (may be just one option of several) , and the use of lidar ratios of 88sr, 75sr, and 55sr for heights above 2.5km is just playing around with lidar ratio values, nothing else.

The analysis of the effects due to the use of altitude independent lidar ratios has been approached in a totally different way in the revised manuscript, as it is outlined in the following. In the new approach, the whole aerosol layer has been divided in two “selected” aerosol layers which are supposed to be characterized by different mean lidar ratio values at each lidar wavelength. Then, it has been allowed the AOT of each aerosol layer to vary of a given percentage, by keeping unchanged the AOT of the whole aerosol layer. The inversion of the lidar signals trough the implemented iterative procedure has been carried out for each aerosol layer. As a consequence, the lidar ratio values retrieved from the implemented iterative procedure for one layer are different from the ones of the other layer. Then, Angström coefficients and differences have been evaluated for both layers to investigate the effects of using different LRs for the two layers at 355 nm, 532 nm, and 1064 nm, respectively.

Page 15, from line 23 to the rest: I stop reviewing the rest of the paper. Again, the scientific value is rather low, most remaining parts contain speculations only. We need a robust uncertainty analysis! This is the main message of this review. Without that, the paper has to be rejected.

We believe that a robust uncertainty analysis will be provided in the revised manuscript as the excerpt of Section 2.2 reported above allows inferring. In addition, a large effort has been done in the revised manuscript to the better demonstrate that depolarization lidar measurements, analytical backtrajectory pathways, dust particle concentration profiles from the BSC-DREAM model (www.bsc.es), satellite images, and AERONET sunphotometer measurements collocated in space

and time with lidar measurements allow to support main results retrieved from the used classification framework.