Dear Referee #3,
many thanks for your comments which have allowed to improve the paper. We have done our best to fulfil the paper's weakness in the revised manuscript as it is highlighted in the following.

The authors present a graphical method that is used to derive particle size and the mixing-ratio of the fine-mode and coarse-mode particles. The input data are vertically resolved (profiles) of extinction coefficients derived from simple backscatter lidar measurements. The authors present results of several case studies of data taken with lidar in Lecce, southeast Italy. The authors' mains conclusion is that it is important to understand the fact that particle properties may change with altitude and that Lecce experiences various pollution conditions like dust from Africa, pollution from the Mediterranean region and long-range transport from North America. The authors conclude that their method is applicable for future research. The paper is not acceptable. It lacks in novelty, ….

The novelty of the paper is due to the use of the **graphical method** of Gobbi et al. (2007) to estimate changes with altitude of aerosol properties by using multi wavelength lidar measurements. The graphical method of Gobbi et al. (2007) which was proposed for the first time on 2007 has been applied to AERONET data. On the contrary, it has been applied (for the first time, to the best of our knowledge) to multi wavelength lidar measurements in the submitted manuscript.

We know that vertically resolved Angström coefficients (å) are commonly used in the lidar community to infer the dependence on altitude of aerosol properties. The use of **a classification framework** to visually convert å and its spectral curvature (Δå) to both the fine mode aerosol radius and the contribution of the fine mode aerosol to the AOT represents the novelty of the results presented in the submitted manuscript. We believe that this technique could be of interest to the lidar community when lidar signals at three wavelengths are only available.

……..has strong weaknesses in the methodology,….

An improved methodology has been used in the revised manuscript. Systematic uncertainties from different sources have been taken into account in the revised manuscript. More specifically, we have allowed the input parameters required 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 Marenco 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 manuscript is reported below for your convenience:

" 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 Marenco 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 $\beta_T(z_f) \cong \beta_M(z_f)$. 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 5×10^{-7} (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 AOT$_{1,ref}$. 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 trough 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 (AOT$_{1,ref}$) 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 AOT$_{1,ref}$. 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 trough 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"

……….lacks any kind of convincing comparison, ….

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 were used to support main results retrieved from the used classification framework.

……………..and the results largely repeat findings from the multitude of lidar observations taken in EARLINET (south European stations) that show similar findings…..

This seems like a good news, because this means that reliable results were presented in the submitted manuscript in spite of strong weaknesses in the methodology. The main point of the paper is to present a method to obtain information on aerosol size distribution with elastic signals only.

I acknowledge that each EARLINET station in itself adds a valuable piece of information to the overall knowledge that is collected on aerosols, their spatial and temporal distribution, and particular properties at each station along the Mediterranean rim. So each station certainly bears its own merit in this network and should not be neglected only because similar findings were made by another station. It is the statistics that counts, and by adding similar observations from different stations this statistics can be collected as added value. This is a good thing to do.

We share your last comments.

……However, I do not see the novelty of work in this paper, as it is incomplete, and the applied methodology itself introduces errors that lead to wrong or biased conclusions.
In summary: the robustness of this study is not presented in any convincing manner.
It is known that the vertical dependence of aerosols exists…

We believe that the above reported excerpt of Section 2.2 highlights the robustness of the study reported in the revised manuscript.

………..The graphical method the authors use is not that new at all. In that regard I am missing reference to the papers by o'Neill (Applied Optics, 40, 2368-2375, 2001; JGR, 106, 9787-9806, 2001; maybe even JGR 107, EID = 4125, 2002 may be relevant in this context). In view of these reference, even though they may not 100% reflect the current approach, I am wondering about any other publications that may also be missing in the reference list, which in turn does not allow me to understand the novelty of this present paper. Coming back to o'Neill: he showed long time ago that the curvature of the extinction spectrum can be used to extract information on the fine-mode and coarse mode fraction of aerosol. So that would leave me with the novelty (in this present paper) that vertical profiles of extinction have never been analyzed in that way, and as far as I recall there may have already been a previous paper on this present method being published some time ago. The authors may want to comment on this, as I may be wrong in my assumption…….

Both the paper of Schuster et al. (2006) and the one of 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. An overview of the publications related to the Angström coefficient and its curvature by O'Neill and other authors has been reported in Schuster et al. (2006) and Gobbi et al. (2007), where the work of Schuster et al. (2006) was also accounted for. As a consequence, we thought that it was not

necessary to mention the O'Neill works and the ones of other authors in the manuscript, even if we were certainly aware of the O'Neill publications. However, the paper by O'Neill et al. (2003) has been added to the references of the revised manuscript. As it has previously been told the graphical method of Gobbi et al. (2007) was proposed for the first time on 2007 and then, it was applied to AERONET data by other authors (e.g. Basart et al., 2009). It has been applied to multi wavelength lidar measurements for the first time in the submitted manuscript, to the best of our knowledge.

…….The main flaw is that the authors wash away the fact that they do not use Raman lidar or HSRL to measure extinction which provides the necessary precision of the extinction data so that the graphical curvature method can be used in a robust way. I do not mean to say that every lidar in the world must be Raman lidar or HSRL, but if a lidar has no Raman channels any lidar team these days must put specific emphasis on error analysis and carefully consider conclusions in view of lack of robustness of extinction data. The authors use backscatter profiles, make assumption on the lidar ratio (in part corroborated by backward trajectories), they extract extinction profiles, and then use the curvature method to obtain their results on mixing ratios of fine-coarse mode, and particle size (or mean size). The authors in a very unclear manner make a sensitivity analysis in which they claim that the errors of the extinction values are not that much influenced by their assumptions, though they have to make quite a bit of guess work on the lidar ratios. Such conclusions as drawn in this paper simply cannot be made on the basis of simple backscatter lidar ………..

We believe that the improved methodology used in the revised manuscript where systematic uncertainties from different sources have also been taken into account, has enabled us to significantly decrease the weaknesses of the work.

……….and the fact that lidar ratios vary with height, even more if we consider that lidar ratios cannot be measured with 0% measurement error. Even 10% measurement uncertainty are hard to achieve with Raman lidar; Angstrom exponents also do not have errors less than 20% in that case. Experience with Raman lidar (which measures extinction) rather shows errors of 30-40% and I do not see these error bars in figure 2b, 2d, 5b, 5d. I do not find a clear description (in terms of hard numbers that can be clearly followed) of error-propagation of this complex analysis. Error bars are missing in spots where it is crucial to show them. Figures 8 and 9 are prime examples of avoiding error bars, and this gives a false impression on the accuracy (and precision) of the method…..

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.

The corroboration by backward trajectories has flaws. For instance, the trajectories in figure 4a show advection from North America. When I counted the symbols in this 2-d plot and compared to the lower panel (Figure 4b) it seems that more symbols (time steps) are shown. So some part of the trajectories in Fig. 4a is missing. This in turn makes me wonder what I am seeing in this trajectory plot, and if the conclusion of

long-range transport is justified at all.

Sorry, some part of the trajectories in Fig. 4a was missing, as a consequence Fig. 4a has been replaced in the revised manuscript.

……………We need to keep in mind that the trajectories need to be somewhat close to the ground which in turn means that the respective air parcels could have picked up aerosols which then might have been transported over long distances from the US to Europe. Was this the case here, or couldn't it be the case that the air parcels picked up aerosols in the Mediterranean area, just 1 or 2 days prior to arrival over Lecce?

The discussion on the back trajectory pathways has been improved in the revised manuscript. An excerpt of Section 4.1 of the revised manuscript where the 28 July-backtrajectory pathways are presented, is reported below for your convenience:

"…………….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. It simulated for the monitoring site of this study, the existence of a dust layer centred around 1.7 km at 12 UTC. Figure 2c (red line) shows the vertical profile of the dust particle concentration from the BSC-DREAM model (http://www.bsc.es/earth-sciences/mineral-dust-forecast-system/bsc-dream8b-forecast/north-africa-europe-and-middle-ea-0) which reaches the value of ~ 25 $\mu g/m^3$ at 1.7 km a.g.l………
………The $\eta(z)$ and $R_f(z)$ values within 3.2-4.8 km a.g.l. are likely due to anthropogenic pollution lifted up to altitudes $z > 2$ km a.g.l. and then transported over south eastern Italy, in accordance with Fig. 4."

*……………….. So is the conclusion on aerosol types, which decides on
the correct choice of lidar ratio, which in turn decides on the correct extinction values
and their uncertainties correct at all? In the end I have to state that this whole approach
comes very close to a circular argument……*

We would like to stress that lidar ratios and corresponding uncertainties are essentially determined by co-located optical thickness measurements in the revised manuscript. So, the new methodology does not lead to a circular argument.

*The AERONET data in that regard do not prove a lot, as they are describing the columnar
results, but the novelty of the paper can only hold for applying the method to the
aspect of analyzing vertical profiles.*

O.K. However, we believe that the reliability of paper results may be supported by AERONET aerosol products if they are within the variability range of the corresponding vertically resolved data.

I also see flaws in data analysis of the lidar profiles. The Angstrom profiles in fig. 2b and 2d show a continuous increase of the Angstrom exponents with height. This increase is so "stable", I have never seen this before in my career. I do not want to argue that only because I have not seen such a behavior it may not be possible that it exists. But I do know that such systematic behavior can easily be produced by wrong alignment of the lidar instrument or a flaw in data analysis: I simply have seen it too many times before, and after considering all possible error sources such kind of profiles returned to what I would like to call "normal" behavior of the vertical distribution of aerosols and Angstrom exponents.

Please note that extinction coefficient profiles as the ones of Fig. 2a are not very common over south eastern Italy and the 28 July, 2011 study case was inserted in the paper for its peculiarity.

The argument that DREAM results support the results of the lidar analysis has flaws in itself. It is known that the dust emission models still are not accurate enough to provide an unambiguous proof of experimental data. Particularly in this present study the model data must be of high quality, because the lidar data simply do not have the necessary robustness. What also strikes me is the fact that figures 2c and 12b show model results (DREAM model) with a comparably high dust concentration. Why is the volume depolarization ratio less than 5%? Even if I consider a reasonable conversion to particle depolarization ratio I do not see a way of ending up with values of more than 10% for the particle depolarization. In view of the overall set of data presented in this study, 10% particle dust depolarization (even under consideration of mixing conditions) cannot be true. Either this number for the depolarization ratio (5% or less) is a typo (I also see low depolarization ratios in Fig. 5a) I am not able to understand the results at all.

Particle depolarisation ratios will be provided in the revised manuscript and it will be shown that the particle depolarization reaches a peak value of about 25% on 5 September, 2011. To this end, it is interesting to note that the dust concentration from the BSC-DREAM model reached a peak value of about 100 μgm^{-3} and of about 25 μgm^{-3} on 5 September, 2011 (Fig. 12b) and 28 July, 2011 (Fig. 2c), respectively.

I find additional flaws in data analysis, but I think I made my point clear. The methodology certainly bears its advantages, however neither the way of data analysis nor the quality of data is good enough to draw the kind of conclusions made in this paper. Last but not least: the authors strip themselves off the merit of the paper because they show three cases studies, nothing more.

Actually, apart the three case studies, results from a total of 11 cases have been reported.

…………………. In view of long-term studies at this site a statistical analysis could extract problems with data analysis and also add valuable information to our overall knowledge of the vertical distribution of aerosols in South Europe.

Demonstrate the reliability of the proposed methodology was the main goal of the submitted paper. We will consider your suggestion in the future if the paper will be accepted, as we hope.

I recommend that the authors withdraw this paper and start all over again. They should

reanalyze their data, include a comprehensive and traceable error analysis, give credit to publications related to their methodology, search for similar studies so that they can corroborate the novelty of their work, and push their work to a more comprehensive description of the aerosol situation in South Italy.

Data have been reanalysed, a comprehensive and traceable error analysis has been included, and credit to publications related to the methodology has been given in the revised manuscript, in accordance with your last suggestions.