The title of the paper may suggest a more general study, not just based on two cases only. That should be made more clear, may be even in the title.

The title has been changed. The new title is “Study of hygroscopic growth of atmospheric aerosol particles using active remote sensing and radiosounding measurements: selected cases at Southeastern Spain”

However, why is that paper not sent to ACP? It is a paper on application rather than on the development of a new measurement technique

The manuscript was first sent to ACP, but it was considered out of the scope of the journal and suggested to be resubmitted to AMT.

Details:

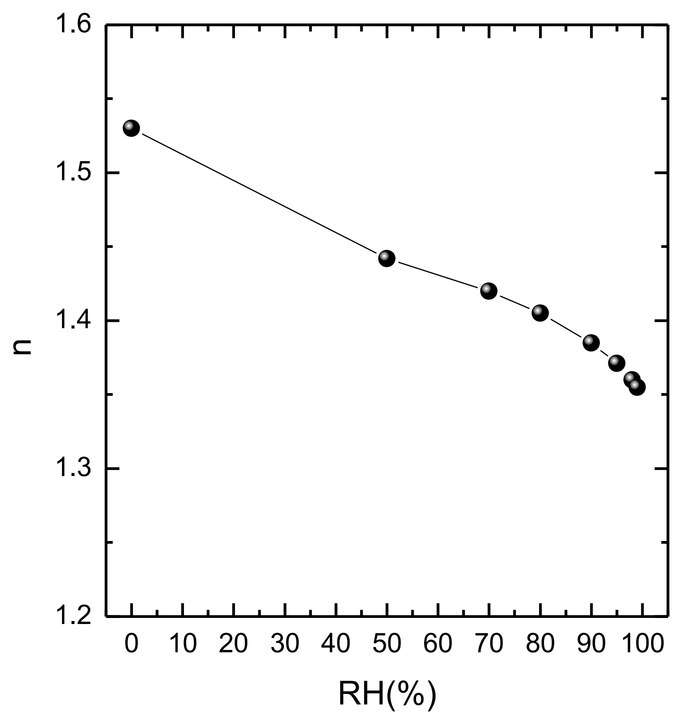
P10294, L12: Please provide information for the relative humidity (40% to 80%, 60%to 90%,:::?) for the enhancement factors of 2.10 and 3.90.

This information was already included in Lines 12-13: “*Values of the aerosol particle backscatter coefficient enhancement factors range from 2.10 ± 0.06 to 3.90 ± 0.03, in the ranges of relative humidity 60-90% and 40-83%, being similar to those previously reported in the literature.*”

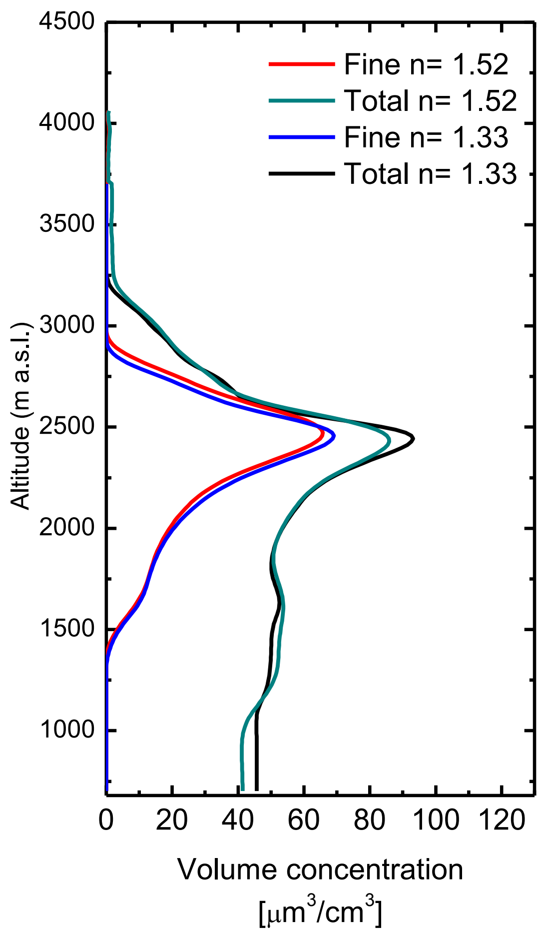
P10294, L24: In the retrieval of the volume concentration the refractive index must be assumed, and the refractive index changes with increasing amount of water in the particles towards the refractive index for water. Is that considered in the study? Did you check the resulting uncertainty by neglecting this effect?

The complex refractive index in LIRIC is not assumed, but is taken from AERONET data. In our case, the values used are 1.33-0.0035i and 1.43-0.0035i at 440 and 1020 nm respectively for Case I and 1.44-0.008i and 1.48-0.008i at 440 and 1020 nm for Case II.

The main inconvenient with LIRIC regarding the refractive index is that is assumed as height independent and at this point we agree with the reviewer that this is an uncertainty source. However, since LIRIC is not an open code we cannot introduce a height-dependent value of the refractive index in order to calculate a reliable value of the uncertainty. Nonetheless, we performed some tests with LIRIC by varying the refractive index provided by AERONET in the input data and keeping all the other parameters in LIRIC code unvaried. In order to try to stablish the maximum possible variation of the refractive index with relative humidity, OPAC software [Hess et al., 1998], based on Mie calculations has been used. An aerosol composed of only water soluble particles, which will present the maximum variation of the refractive index in case of hygroscopic growth, has been assumed to determine the variations of the refractive index with relative humidity. Values of the real part vary between 1.35 at a relative humidity close to 100% and 1.52 at RH = 0%, as we can see in the figure below.



According to LIRIC retrieval, using values of the real part of the refractive index of 1.35 and 1.52, the volume concentration profiles retrieved for the different values were almost the same (with differences below 7.5%), as we can see in the figure below, corresponding to the case on 22 July 2011. Variations in the imaginary part made no difference in the output results and they are not shown in the plot.



Therefore, it can be assumed that LIRIC's sensitivity to variations of the refractive index is very low and the associated uncertainty is quite small. In addition, in the range of relative humidity we are working in the study, values of the refractive index range between 1.38 and 1.45 according to OPAC and, in addition, we do not have an aerosol mixture composed of only water soluble elements. Therefore, variations of the output results in our case studies will be much lower than the ones presented here.

Nonetheless, we agree with the reviewer and a more exhaustive analysis, with a larger number of cases, would be necessary in order to reliably quantify the uncertainty due to the refractive index height-independency assumption in LIRIC for future studies.

Additional information regarding the refractive index has been included in lines 201-215:

"It is worthy to point out here that the refractive index, percentage of sphericity and size distribution for each mode are assumed as height-independent in LIRIC retrievals. In cases of different aerosol types in the atmospheric column or variation of these properties with height, this assumption may introduce some uncertainties. Therefore, it is necessary to consider the limitations of LIRIC under these specific situations when analyzing LIRIC results. Ancillary information, such as the lidar retrieved optical properties profiles, has to be used in order to guarantee the reliability of the results. In cases of hygroscopic growth, it is clear that the refractive index is not height-independent, since it changes with changing relative humidity. However, we performed some tests with LIRIC by varying the refractive index provided by AERONET, which is the one used by LIRIC, and keeping all the other parameters in LIRIC code unvaried. The volume concentration profiles retrieved for the different values of refractive index were almost the same (with differences below 7.5%). Therefore, it can be assumed that LIRIC's sensitivity to variations of the refractive index is very low and the associated uncertainty is quite small."

Hess, M., Koepke, P., & Schult, I. (1998). Optical properties of aerosols and clouds: The software package OPAC. *Bulletin of the American meteorological society*, *79*(5), 831-844.

P10300, L5-15: The procedure to obtain the volume concentration profile must be explained in more detail. Sun photometer data are probably inverted by assuming a fixed (height-constant) set of refractive index parameters. But in the vertical, the true refractive index is continuously changing. So, the assumption of a height-independent refractive index causes errors in the LIRIC approach as a whole. The consequences for the retrieved volume concentrations must be discussed. We need a discussion on the uncertainty here. Changes in the volume concentration with increasing relative

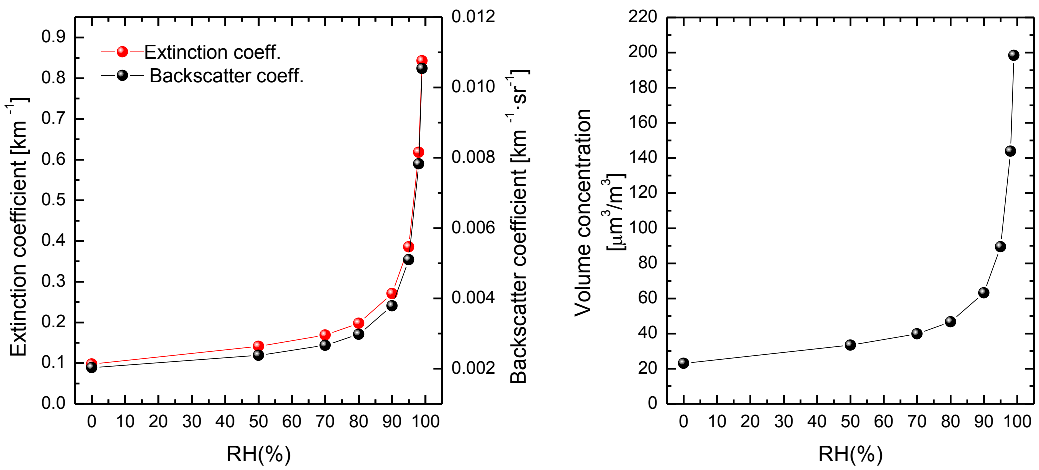
humidity may just be the response to the erroneous refractive index assumption. On the other hand, it should be possible (by using a Mie scattering model), to check whether the increase in particle backscatter and extinction coefficient is consistent with the increase in volume concentration

In order to clarify this point, more details about LIRIC are provided in lines 189-201:

"*From AERONET code, input variables used by LIRIC are columnar integrated volume concentration values for each mode, refractive index, single scattering albedo, integrated backscatter coefficients, the first and second diagonal elements of the scattering matrices at 180º, the fraction of the spherical particles and the aerosol optical depth. In addition, based also on AERONET code, an aerosol model, defined by the columnar integrated volume concentrations of each mode (fine and coarse modes) and based on a mixture of randomly oriented spherical and spheroid particles, is assumed [Dubovik and King, 2000; Dubovik et al., 2006]. These data, together with the elastic lidar raw signals at 355, 532 and 1064 nm, are used to obtain the volume concentration profiles for fine and coarse particles, distinguishing between coarse spherical and coarse spheroid mode when 532-nm cross-polarized lidar channel is used. The mathematical procedure used to retrieve the volume concentration profiles is described in detail in Chaikovsky et al., [2008].*"

As it was explained in the previous response, LIRIC's sensitivity to variations in the refractive index is quite low. Therefore, even though an error is introduced by LIRIC when assuming a height-independent set of refractive index values and the uncertainty needs to be taken into account, the noticeable changes in the volume concentration with increasing relative humidity obtained here are much larger than the retrieved uncertainty and cannot be just explained as a LIRIC artifact.

The increase in the volume concentration with relative humidity is also observed if we perform Mie calculations by means of OPAC. As it can be seen in the figure below, assuming a continental polluted aerosol type similar to the one we presented in our case studies, an increase in the aerosol extinction and backscatter coefficients with relative humidity is obtained. This increase is simultaneous to the increase in the volume concentration, which is in agreement with our experimental results.

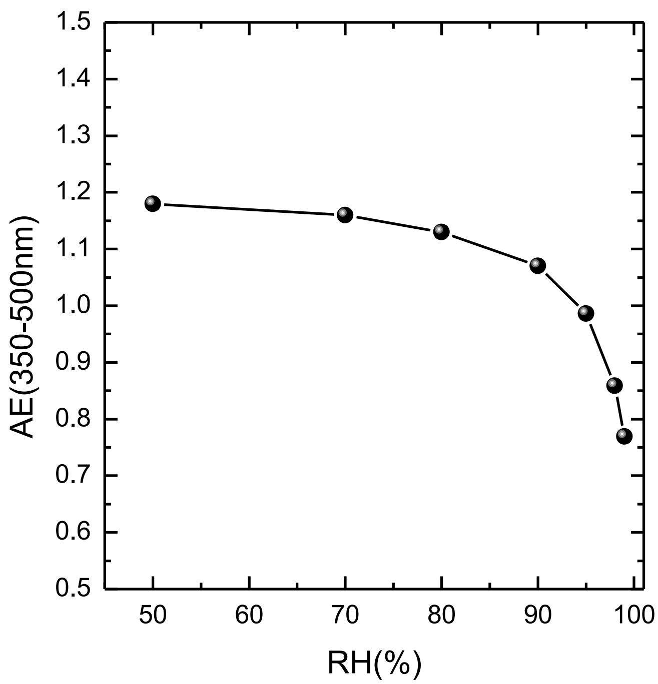


P10301,L1: Why is a simultaneous decrease of the beta AE and depol. ratio an indication for larger and more spherical particles? Did you perform Mie scattering modeling to show how large the effect on the Angstroem value is when the size distribution is slightly changing (shifted) by water uptake. I believe this is necessary to show that based on modeling, because my feeling (no knowledge) is that the changes in the size distribution are so small that the response of the wavelength dependence of the optical properties is almost not visible.

Furthermore, why should the particles (here dust) become more spherical? If they are hydrophobic, nothing will change with increasing relative humidity. And even if they attract some water, will that really change their shape. So, please keep the speculative discussion a bit more open for other possibilities.

Smaller values of the AE indicate a predominance of larger aerosol particles in the aerosol mixture [Dubovik et al., 2002]. Also, linear particle depolarization ratio is related to the particle shape, with lower values corresponding to more spherical particles [Sassen et al., 1991; Schotland et al., 1971]. Therefore, a simultaneous decrease on both is related to an increase in the presence of larger particles in the aerosol mixture and variations to more spherical particles. In cases of hygroscopic growth the aerosol tends to increase its size and to solve in water acquiring a more spherical shape [Inerle-Hof et al., 2007]

OPAC software, based on Mie calculations, has been used in order to analyze the influence of water uptake on the Angström exponent, assuming a continental polluted mixture aerosol type. As we can see in the figure below, the variations in the extinction-related AE(350-500nm) are quite small, ranging between 1.18 and 1.07 for relative humidity between 50 and 90%, similar to what we obtain in our experimental results. Larger variations are obtained for values of the RH larger than 90%, which are not observed in our study.



In the case studies presented here, mineral dust is not observed. Only in case I, some traces of remaining dust from a previous dust event were observed, located mainly at higher altitudes than the analyzed layer. Text has been reviewed and modified accordingly to avoid confusion at this point. According to our results, the aerosol mixture is hygroscopic, not hydrophobic, and as previously stated, in cases of hygroscopic growth the aerosol tends to solve in water acquiring a more spherical shape.

Sassen, K.: The Polarization Lidar Technique for Cloud Research - a Review and Current Assessment, B Am Meteorol Soc, 72, 1848-1866, 1991.

Schotland, R. M., Sassen, K., and Stone, R.: Observations by Lidar of Linear Depolarization Ratios for Hydrometeors, J Appl Meteorol, 10, 1011-1017, 1971.

Inerle-Hof, M., Weinbruch, S., Ebert, M., & Thomassen, Y. (2007). The hygroscopic behaviour of individual aerosol particles in nickel refineries as investigated by environmental scanning electron microscopy. *Journal of Environmental Monitoring*, *9*(4), 301-306.

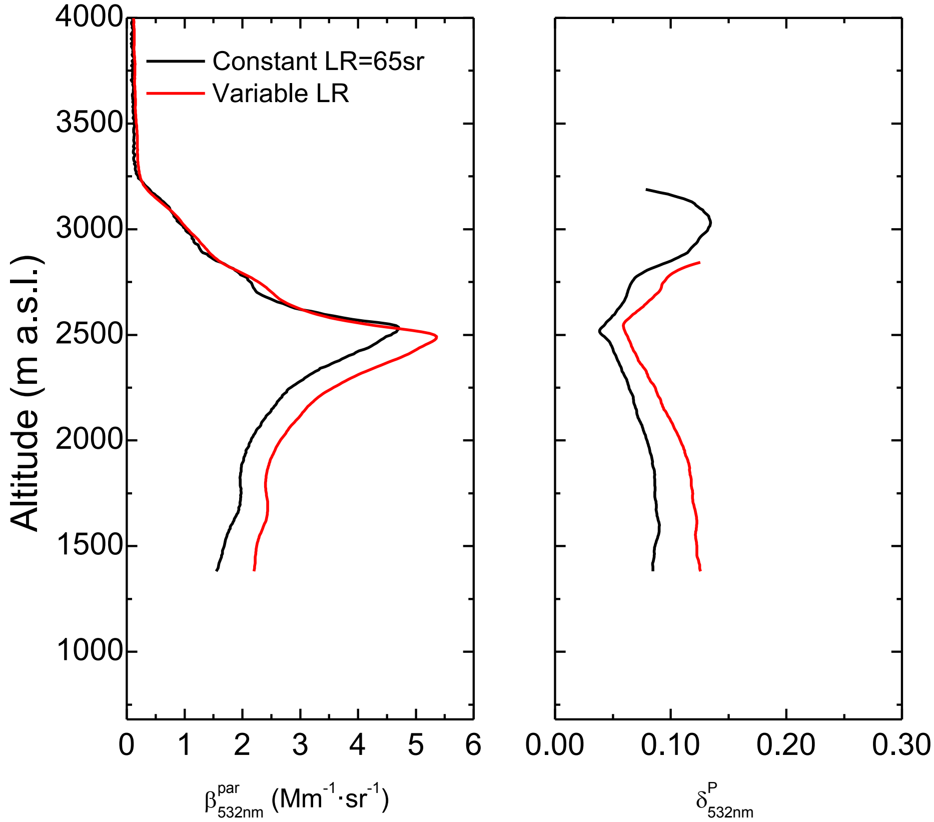
May be your retrieval of the particle backscatter coefficient (with fixed, height-indendent lidar ratio) is erroneous. The error is then also a function of height, the error is stronger for 355nm than for 532nm and changes with height when using the Klett algorithm!And this height-dependent error shows then up in the Angstroem retrieval, but also in the retrieval of the particle depolarization ratio (which is a function of the retrieved backscatter coefficient)! So, at the moment, for me it is not clear whether we see a clear atmospheric effect or whether the found features are just introduced by errors in the backscatter and depolarization ratio retrieval!

The assumption of an independent-range lidar ratio (LR) is the simplest way to retrieve the backscatter coefficient profile. This is a source of errors, but even incorrect assumed backscatter-to-extinction ratios can yield rather accurate results [Kovalev 1995]. In case of profiles that include multiple aerosol layers with different properties, the assumption of an independent-range LR can cause some distortion to the retrieved backscatter coefficient profile. For a given aerosol layer, the distortion-associated error depends on both the backscattering ratio and the differences between the real LR and the assumed one [Kovalev and Eichinger, 2004]. As indicated by the reviewer, the influence of the error is higher at shorter wavelengths. This spectral dependence is linked to the strong contribution of the molecular signal to the total signal at short wavelengths [Takamura and Sasano, 1987]. In general, the errors in the profiles obtained with the Klett method considering a constant lidar ratio are less than 20% for the aerosol backscatter coefficient and less than 25% for the linear particle depolarization ratio [Franke et al., 2001; Alados-Arboledas et al., 2011;

Preissler et al., 2011]. Values of the uncertainty in Klett retrieval were used to obtain the estimated errors in the backscatter coefficient enhancement factor shown in Table 2. Since we have calculated new errors for the aerosol particle backscatter coefficient according to the reviewer suggestion, the errors of $f_{\beta}(RH)$ has been recalculated accordingly.

For the different cases of aerosol hygroscopic growth we have analyzed at our station, in general we observed a decrease of the AE with the increasing relative humidity. However, the decrease in the AE is so small that is within the uncertainty of the derived β-AE. As explained in the previous response, this is also obtained when using OPAC to perform Mie calculations. Therefore, a more detailed analysis with more case studies and based on Raman analysis will be needed to come to this conclusion. According to the reviewer's comment, we have reduced the emphasis given to this hypothesis and introduced a more open discussion at this specific point.

However, in the case of the depolarization the decrease is not due to the errors in the retrieval due to the assumption of a height independent lidar ratio. Despite this assumption introduces an uncertainty, the decreasing trend is always obtained, as we can see in the next figure, where two retrievals are presented. The first one is obtained with a fixed lidar ratio (65 sr). The second one is obtained by using a height-variable lidar ratio in the Klett algorithm according to the one proposed in Ackermann et al., [1998] for continental aerosol type, which is increasing almost linearly with relative humidity in the range of RH we are considering.



Alados-Arboledas, L., et al. (2011), Optical and microphysical properties of fresh biomass burning aerosol retrieved by Raman lidar, and star- and sun-photometry, Geophysical Research Letters, 38, doi:10.1029/2010GL045999.

Franke, K., et al. (2001), One-year observations of particle lidar ratio over the tropical Indian Ocean with Raman lidar, Geophysical Research Letters, 56, 1766{1782, doi: 10.1029/2001GL013671.

Kovalev, V. A. (1995). Sensitivity of the Lidar Solution to Errors of the Aerosol Backscatter-to-Extinction Ratio - Influence of a Monotonic Change in the Aerosol Extinction Coefficient. Applied Optics, 34(18), 3457-3462.

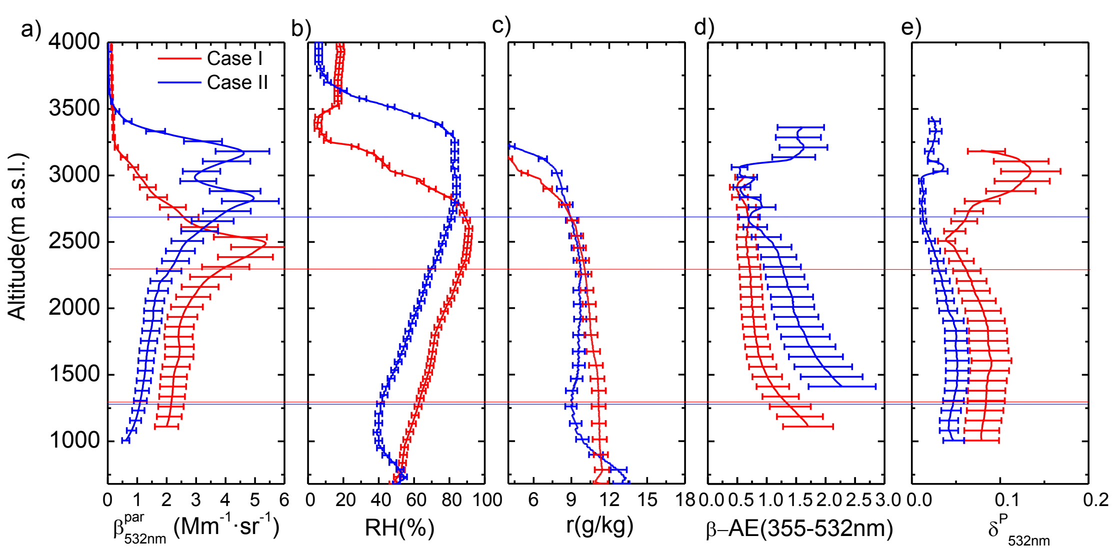
Kovalev, V. A. and W. E. Eichinger (2004). Elastic Lidar: Theory, Practice, and Analysis Methods, Wiley.

Preissler, J., F. Wagner, S. N. Pereira, and J. L. Guerrero-Rascado (2011a), Multiinstrumental observation of an exceptionally strong Saharan dust outbreak over Portugal, Journal of Geophysical Research, 116, D24,204, doi:10.1029/2011JD016527.

Takamura, T., & Sasano, Y. (1987). Ratio of aerosol backscatter to extinction coefficients as determined from angular scattering measurements for use in atmospheric lidar applications. Optical and quantum electronics, 19(5), 293-302.

P10301, L29: The height-independent water vapor mixing ratio obtained with radiosonde is the most important quantity to convince the reader that you detected one and the same aerosol mixture, and that changes in the aerosol mixture with height are the response to water uptake. Thus in the figures, I would show the optical and microphysical properties always together with the radionsonde water vapor mixing ratio profile, not only with the relative humidity (as done in Figure 3). Both parameters should be shown in Figure 3.

Water vapor mixing ratio profiles has been included in Figure 3.



"Figure 3. a) β^{P}_{532nm} retrieved with Klett-Fernald algorithm (LR=65 sr for Case I and LR=70 for Case II) from 20:30 to 21:00 UTC on Case I and 20:00 to 20:30 UTC on Case II b) RH profiles from the radiosounding launched at 20:30 UTC on Case I and at 20:00 UTC on Case II. c) *r* profiles from the radiosounding launched at 20:30 UTC on Case I and at 20:00 UTC on Case II. d) β-AE(355-532nm) retrieved with Klett-Fernald algorithm for the same periods. e) δ^{P}_{532nm} retrieved from lidar data for the same periods. Horizontal lines represent the height limits of the aerosol layers selected for the analysis in Case I (red line) and Case II (blue lines)."

P10303, L6: So, here we introduce how important the chemical composition is and that it changes with water uptake, which then will change the refractive index characteristics. That must be clearly stated. Please provide a consistent picture.

According to previous responses, it has been clearly stated in Lines 208-210:
"In cases of hygroscopic growth, it is clear that the refractive index is height-dependent, since it varies with increasing/decreasing relative humidity due to the changes in the aerosol composition."

P10305, L22 Again, the Klett-Fernald procedure.: although the relative humidity is continuously increasing, water uptake takes place, chemical composition, refractive index, and size distribution changes, you select height-independent lidar ratios of 65 and 70sr (in agreement with AERONET observations, performed hours before). Did you check by Mie scattering calculation whether the impact of relative humidity is low on lidar ratio, but still significant for backscatter Angstroem exponent? May be check the paper of Ackermann, 1998 (J AO Tech?). He computed some lidar ratios as a function of relative humidity. However, if the particles are absorbing (and your lidar ratios of 65 to 70sr point in this direction) the lidar ratio may drop to values for non absorbing particles (around 40sr) when they attracted a lot of water so that the water fraction dominates when the relative humidity is close to80% and higher. So, there are many sources that introduce uncertainties in the entire approach. With the wrong backscatter coefficients (because of wrong lidar ratio profile) you may get wrong Angstroem exponents and wrong particle linear depolarization ratios.

We performed Mie calculations by using OPAC software. As we can see in the figures below, there is a variation around 10 sr in the LR for a continental polluted aerosol type in the range of RH we are studying in this paper. Considering this and the information in Ackerman’s study, we performed the test previously explained. We considered the assumption of a constant lidar ratio as a source of uncertainty and it is included in the error bars in figure 3, assuming errors around 20% for the aerosol backscatter coefficient and 25% for the linear particle depolarization ratio [Franke et al., 2001; Alados-Arboledas et al., 2011; Preissler et al., 2011]. For the Angström exponent the variation is below 0.2, according to OPAC output values. The variation of the Angström exponent is similar to what we obtained in our experimental results, as previously explained. However, since it is within the uncertainty limits, the emphasis to this hypothesis has been reduced in the text and is clearly explained so that the reader is aware.

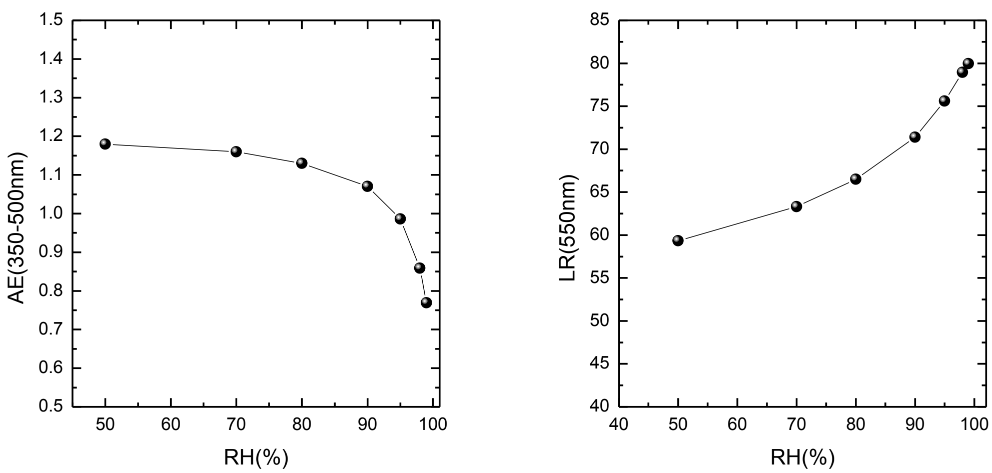


Figure 1:

Why do you show single scattering albedo, more useful would be the refractive index (real part and imaginary part)?

The single scattering albedo is shown here mostly to characterize the different aerosol types in the column during the analyzed days. Both the single scattering albedo and the refractive index provide similar information regarding this point. The single scattering albedo is presented here because is more widely used in the literature and comparison with previous studies is easier in order to characterize the aerosol types [Dubovik et al., 2006]

The AERONET time series (a, b) indicate changing aerosol conditions after 1800 (UTC?). Do you have lidar measurement before sunset (may be around 1600UTC), to estimate the change in AOT from, e.g. 1600UTC to 2000UTC, based on lidar (that would support the use of AERONET data)?

Unfortunately, there are no additional data from 1600 UTC. Since atmospheric conditions after sunset are more stable due to the decrease in the convective activity, we have assumed that the use of AERONET data from late evening using lidar data at 2000 UTC, which is only 2 hours difference, is not a source of large uncertainty. Nonetheless, we have compared the aerosol properties during late evening on both 22 July 2011 and 22 July 2013 with the aerosol properties during the early morning on 23 July 2011 and 23 July 2013 and they present almost identical values for the Angstrom exponent. For the aerosol optical depth some small changes are observed, especially in case II. However, we have to take into account that these changes in the aerosol load took place in a period of ~10 hours.

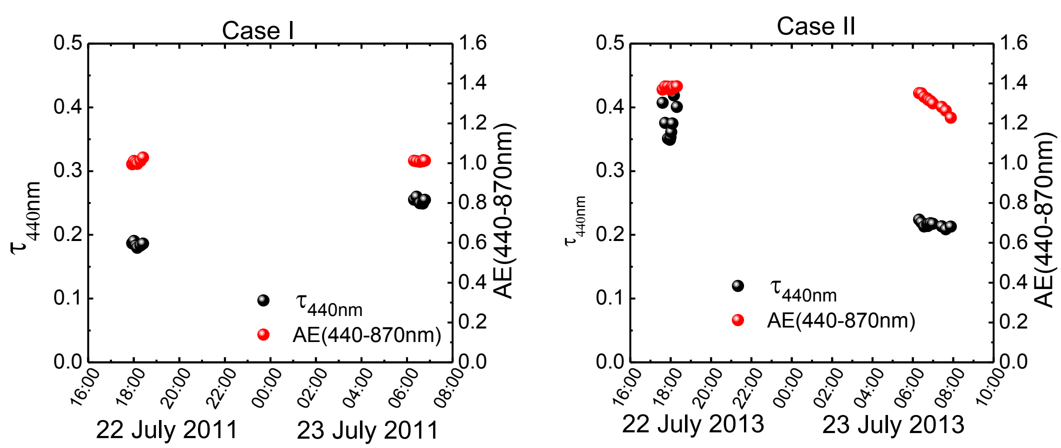
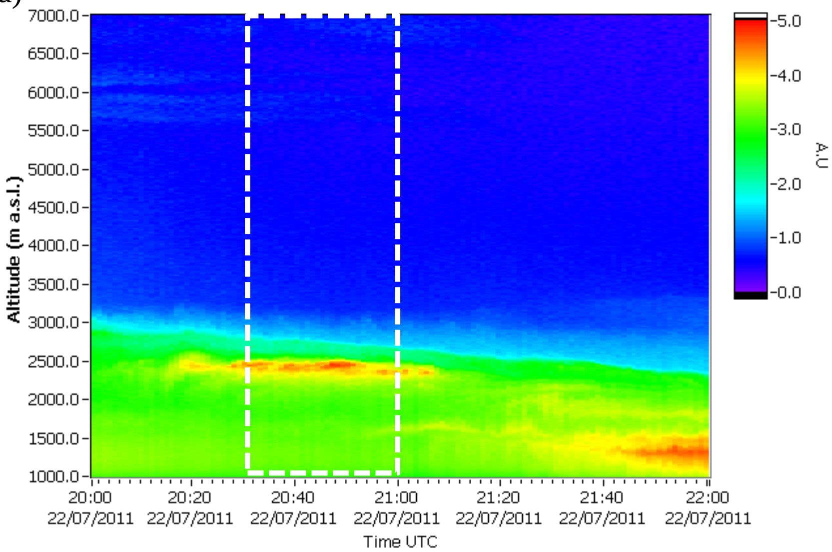


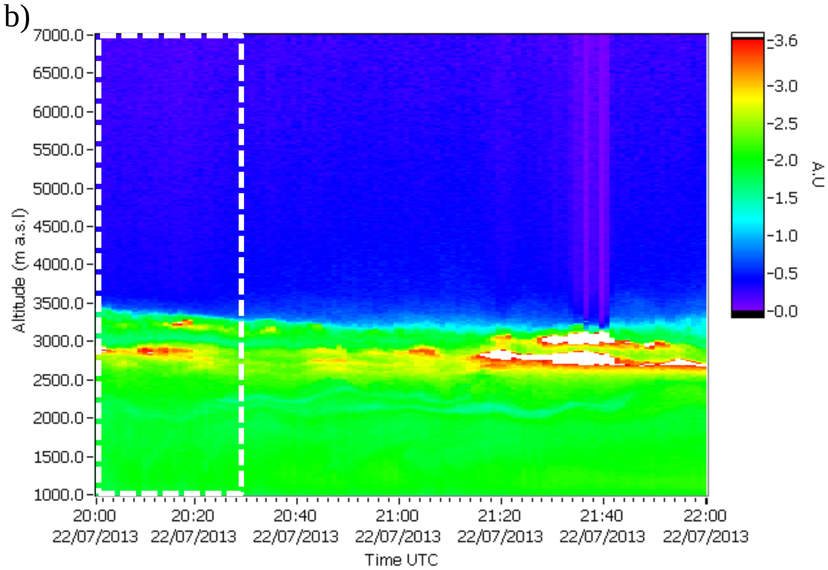
Figure 2: If you have lidar observations over the entire day, please show them and indicate the data analysis period by white vertical lines. Please state in the caption that all features in the color plots are aerosol related (top), or if clouds were present (bottom), and also state that you cloud-screened the data before used in the investigations of the water uptake effect, in the caption.

As previously said, there are no additional data during these case studies. White vertical lines have been added to indicate the analyzed periods and the caption has been modified according to the reviewer's suggestions.

a)



b)



"Figure 2. a) Lidar RCS time series at 532 nm (arbitrary units) on 22nd July 2011 from 20:00 to 22:00 UTC. Features shown here are aerosol related. b) Lidar RCS time series at 532 nm (arbitrary units) on 22th July 2013 from 20:00 to 22:00 UTC. For this case, some clouds were present between 21:35-24:45, leading to a strong attenuation of the RCS. On both cases data were cloud-screened before proceeding to the investigation of hygroscopic growth effects. "

Figure 3: I do not believe the small error bars in the backscatter plot (a) and Angstroem plot (c), because of the lidar ratio uncertainty, lidar ratio is assumed in the retrieval to be height-constant, but is probably height variable. After sunset you do not have AERONET AOT values, so even the numbers 65 and 70sr may not describe properly the situation.

As mentioned, show water vapor mixing ratio, too.

As previously stated, water vapor mixing ratio was included in Figure 3b. In the previous version of the manuscript, error bars in the backscatter coefficient were obtained according to the methodology described in Pappalardo et al., [2004] and Guerrero-Rascado et al., [2008], which considers only statistical errors. In order to take into account the errors introduced by the assumption of a constant lidar ratio the errors bars have been modified assuming a relative error of 20% for the aerosol particle backscatter coefficient and 25% for the linear particle depolarization ratio, as indicated in [Franke et al., 2001; Alados-Arboledas et al., 2011; Preissler et al., 2011].

Figure 4: Backward trajectories show that there must also be a strong maritime component in the aerosol mixture. These maritime particles may dominate the observed hygroscopic features. And if the marine particles become partly dry, when the humidity decreases below 50% then they may become (sea salt component) non spherical, and may explain the slightly enhanced depolarization ratio? At least the backward trajectories do not just show the impact of desert dust.

The possible influence of desert dust is mentioned here because of the models used to characterize the aerosol type, which indicated the presence of mineral dust. However, according to our experimental data, at the time we analyzed here only a small residual layer of mineral dust is observed above the analyzed height range, but is not the predominant aerosol type. This presence of mineral dust can be easily explained due to the occurrence of a dust outbreak just before the analyzed period. Nonetheless, in the layer analyzed to study the aerosol hygroscopic growth, the optical properties indicate no presence of dust. The influence of marine aerosol is also considered in the text, in Lines 405-414:

"The backward trajectories analysis also revealed that the air masses were travelling very close to the sea surface above the Mediterranean Sea and marine aerosols might be probably present in the aerosol mixture (Figure 4a). For Case II, the air masses come from the Mediterranean region at the three altitude levels considered (Figure 4b) and they were travelling within the marine boundary layer before reaching Granada station, so they are likely loaded with marine aerosol from the Western Mediterranean Sea together with sulphates and smoke from Europe as indicated by the NAAPS model. The trajectories are very similar to those on Case I, but as for this case they travelled more slowly above the Mediterranean Sea they are likely loaded with more marine aerosol than in the previous case."

However, more emphasis is not given to this hypothesis since the presence of marine aerosol in large concentrations at high altitudes is not usual.

Figure 6: Can we really trust the LIRIC results in these cases of complex mixtures and changing microphysical properties and composition

At least, the discussion should mention the problems, so that the reader can decide what to believe and what is rather uncertain

In cases of aerosol properties changing with height LIRIC presents some limitations, since properties such as the refractive index or single scattering albedo are assumed as constant with height and this is an important source of uncertainty in the results. LIRIC has to be carefully applied in these cases and results need to be carefully analyzed.

However, as previously explained, variations in this refractive index do not introduce large variations in LIRIC output results indicating that LIRIC is more sensitive to some other aerosol properties used in the code, at least in the cases analyzed here. Therefore, results obtained here can be considered as good indicators of the aerosol microphysical properties variations with relative humidity even though uncertainty cannot be neglected and more accuracy will be needed for future studies. A more comprehensive study in collaboration with the developers of the code and including more case studies should be performed to assess this uncertainty.

This information is included in the text in lines 201-215:

"It is worthy to point out here that the refractive index, percentage of sphericity and size distribution for each mode are assumed as height-independent in LIRIC retrievals. In cases of different aerosol types in the atmospheric column or variation of these properties with height, this assumption may introduce some uncertainties. Therefore, it is necessary to consider the limitations of LIRIC under these specific situations when analyzing LIRIC results. Ancillary information, such as the lidar retrieved optical properties profiles, has to be used in order to guarantee the reliability of the results. In cases of hygroscopic growth, it is clear that the refractive index is not height-independent, since it changes with changing relative humidity. However, we performed some tests with LIRIC by varying the refractive index provided by AERONET, which is the one used by LIRIC, and keeping all the other parameters in LIRIC code unvaried. The volume concentration profiles retrieved for the different values of refractive index were almost the same (with differences below 7.5%). Therefore, it can be assumed that LIRIC's sensitivity to variations of the refractive index is very low and the associated uncertainty is quite small."