Reply to #1 referee

The manuscript fits both the journal and special issue scopes, describing an interesting methodology to be applied to existing and/or future ceilometer networks to retrieve vertically-resolved aerosol microphysical parameters. Nevertheless, the paper shows some issues that I am very confident the authors will successfully address, making the manuscript ready for publication.

General Comments

#1 It is very interesting to obtain microphysical aerosol properties using low-cost and low maintenance instruments, however in the paper is missing a discussion on who is going to use the data (end users) and which are end users specific needs, in terms of accuracy. Likewise, it should be also introduced a subsection in the conclusions describing if the proposed approach meets the end user requirements.

- We thank the referee for his/her encouraging comments. As in other AMT-published methodologies (e.g., Veselovskii et al., 2002), there is not a specific "end-user" this work is addressed to. As in that case, we are simply proposing a methodology to retrieve some bulk aerosol properties from lidar measurements, and characterize relevant performances. This methodology can be used in a variety of fields concerning aerosol observations, so we believe it cannot be framed as a deliverable to answer specific "user requirements".
To make our aims clearer, we better specified this point in the manuscript (line 95) in the following way:
"Given the necessity to couple advancement in instrumental technology with tools capable of translating raw data into a robust, quantitative and usable information, we propose and characterize a methodology to be employed in elastic backscatter lidars and automated lidar ceilometer applications to retrieve in a quasi-automatic way vertically-resolved profiles of some aerosol optical and microphysical properties."

#2 The method-associated uncertainty seems to be very optimistic. In fact, the uncertainty is mainly similar or a just a little greater with respect to the highly costly multi-wavelength lidar retrieval, developed by Veselovskii et al., 2002.

- In the work of Veselovskii et al. (2002) the uncertainty associated to volume and surface area retrievals is of 5% for both variables (using a 0.2 µm modal radius PSDs), and 15% and 2% (using a 2 µm modal radius PSDs). This is reported in Veselovskii’s Table 1, input random uncertainty 0%, i.e. the instrumental error condition implicit in our work (our model uncertainty does not include lidar measurement uncertainty). With declared uncertainties of 30-40%, our methodology is 3-8 times less accurate than the two PSD cases addressed by Veselovskii et al. (2002), a result expected from instruments a factor 10-20 cheaper than the ones needed in that case. Furthermore, unlikely Veselovskii et al., (2002), our methodology outcomes are validated against real measurements.
We added a comment on this in the conclusions (line 515): ‘These are higher than those found by Veseloski et al. (2002), applying a method in the retrieval of multi-wavelength lidar systems (10 to 20 times more expensive than ALC systems)’.

#3 The model is based on a priori three-modal log-normal aerosol distribution. Changing the mixing ratio among modes and total particle number impact of course the result. The main
problem linked with this approach, is that the model should be “tuned” on particular atmospheric condition (as for Lecce, where the total number of aerosols have been diminished, hypnotizing cleaner continental aerosols).

- We agree with the reviewer. This aspect is indeed discussed both in the manuscript (section 4.1) and in Appendix D, where we provided a sensitivity test tuning the model to better reproduce aerosol conditions at the Lecce and Potenza sites. We modified a sentence in the conclusion to further stress this point: 'An obvious intrinsic limitation is that the method is dependent on the considered aerosol type and in this study was tuned to reproduce average continental aerosol conditions. Errors associated to the application of the derived functional relationship might be larger if more ‘specific’ aerosol conditions (e.g. contamination by sea salt or desert dust particles) affect a given site.

#4 Simulations from MonteCarlo are fitted with a 7th grade polynomial. Even if in the manuscript follows the approach described in Barnaba et al., papers, no explanation is given about this choice. More important, it is missing in the manuscript a model sensitivity study: how the results are affected, for example if a 3rd degree polynomial is used instead of 7? Is there for example a convergence in the results if the polynomial order is increased? Or is overfitting creating problems? All those question should be properly addressed.

- In the work of Barnaba et al. (2001) a 7th grade polynomial was used because “the polynomial fits need to be extended to the seventh degree to reach a good correlation coefficient (c² > 0.98). See the Appendix A2 of the above-mentioned paper for c² definition.” Similar results have been obtained for this work. Thus, for this reason and for homogeneity with Barnaba et al. (2001) paper, we used 7th grade polynomial. A convergence in the results is observed the polynomial order is increased. A following sentence was added in section 3: ‘The choice of a seventh-order polynomial fit was made for homogeneity with BG01 and BG04a’.

Specific Comments:

#Pag 1 Line 19: Please be more specific about "continental aerosol type". Provide a very short description of the aerosol species belonging to this category.

- Thank you. We added the following sentence: ‘An average ‘continental aerosol type’ (i.e. clean to moderately polluted continental aerosol conditions, e.g., section 2.1) is addressed in this study’.

#Pag 2 Line 43: I suggest to classify aerosol effects on radiation budget into direct, semi-direct and indirect. Moreover, literature should be broaden (e.g. Feingold papers are missing)

-Thank you. We modified the text as follows: ‘Aerosol particles affect the Earth’s radiation budget mainly by two different processes: 1) by scattering and absorbing both solar and terrestrial radiation (aerosol direct effect, Haywood and Boucher, 2000 and aerosol semi-direct effect, Johnson et al., 2004) and 2) by serving as cloud and ice condensation nuclei (aerosol indirect effect, Lohmann and Feichter, 2005, Stevens and Feingold, 2009 and Feingold et al., 2016).

#Pag. 2 Line 54, again literature is poor, I would suggest to add at least Tosca et al., 2017 remote sensing
- Added, thank you for the suggestion.

### Pag.2 Line 57 I suggest to add reference to Lolli et al., 2018 AMT

- Added, thank you for the suggestion.

### Pag.2 Line 59-65 CALIPSO is not the only mission with lidar monitoring the atmosphere from space. Since few years there is also CATS. Please refer to York et al., 2016, McGill et al., 2016

- Added, thank you for the suggestion.

### Pag. 3 Line 85. I suggest to add to the reference Madonna et al., 2018 showing results of the new intercoparison campaign INTERACT-II

- Added, thank you for the suggestion

### Pag. 3 Line 99 and 100: Both measurement units are wrong both for surface area and volume

- Corrected, thank you for this remark.

### Pag. 3 Line 107: please refer to the first comment

- According to this and to a similar comment made by reviewer #2, we reformulated the sentence as follows:

  'we address here an ‘average-continental’ aerosol type (i.e. clean to moderately polluted continental aerosol conditions, e.g., section 2.1), expected to climatologically dominate over most of Europe, despite the not negligible differences that can be encountered across Europe over both the short and the long-term (Putaud et al. 2010)’

### Pag. 4 Line 133: Please be more specific. Now the aerosol type is continental. Which is the difference with "average continental"?

- We now refer to ‘an average continental’ aerosol type (i.e. clean to moderately polluted continental conditions, Hess et al., 1998)’ in the text.

### Pag. 4 Line 139 Eq 1 suffers from hasty writing. Not all the variables are described in the manuscript

- Corrected, thank you for this remark. Sorry for the confusion (see also reply to reviewer #3 on this same aspect).

### Pag. 4 Line 140 rmi is not present in Eq. 1

- Corrected, thank you for this remark.

### Pag. 4 Line 141 Even if it is clear, variables mri and mim are not defined here (fewrows below yes) neither present in Eq.1

- Corrected, thank you for this remark.
(see also our reply above).

**Pag. 4 Line 145** N shows wrong measurement units.
- Corrected, thank you for this remark.

**Pag. 5 Line 176** Hi is not defined. Why on equation 3 is arbitrarily used 5.5 Km? A ref should be at least added.
- Done, thank you for this remark.

**Pag 7 Line 226** Measurement units are wrong.
- Corrected, thank you for this remark.

**Page 11 Line 399**: Please specify which AERONET datasets were used in the manuscript and which version.
- Done, thank you for this remark.

**Pag 13 Line 465**: usually humidity is higher at night

- The referee is right. We report here the reply to #3 reviewer that also had a comment about the impact of RH on the ALC volume retrieval. We know this effect is important in our volume estimates and relevant errors (e.g. also Barnaba et al., 2010; Adam et al., 2012). In fact, the RH dependence is taken into account by the model itself and it is accounted for in the model results variability. Indeed, errors can be much larger in the retrieval of PM loads, where a further unknown (particle density) is involved. In fact, we propose to retrieve volume not mass, and reference aerosol volume measurements are rather complex to perform if not including the full size distribution (as in the case of optical instruments).

A further missing information would concern hygroscopicity of observed aerosols. We believe an extensive discussion (ALC vs other techniques) of volume comparisons would require a full paper itself. We believe it is better here to show some comparisons as in Figure 8 demonstrating the ALC volume estimates can well match the optical ones within the expected relevant variability. However, to provide more information about RH, we added a horizontal bar in the upper part of Figure 8 indicating the range (RH<60%, 60%<RH<90% and RH>90%, respectively) of the measured in-situ RH during the ALC-OPC volume comparison. In this respect, the following text has been added:

This latter effect is confirmed by the large RH values (RH > 90%) measured after 18 UTC. The lower panel shows a good agreement between the ALC-derived and the Fidas OPC Vₘ values, in particular until 04 UTC and after 16 UTC. Some differences emerge around 07 UTC and between 11 and 15 UTC, where the ALC volume is lower by a factor of 2 compared to the in situ Fidas Vₘ values. The smaller minimum detectable size of the Fidas OPC instrument with respect to the OPS is likely the reason for the better accord between ALC and OPC Vₘ values in this test date. For this case, the effect of RH seems to be less important, and indeed RH values keep lower than 90%.

In general, high RH values (RH >= 90%) are known to markedly affect the aerosol mass estimation from remote sensing techniques and its relationship with ‘reference’ PM2.5 or
PM10 measurements methods, usually performed in dried conditions (e.g. Barnaba et al., 2010; Adam et al., 2012, Li et al., 2016, Li et al., 2017). This theme is also discussed in Diemoz et al. 2018a for the ALC measurement site of Figure'.
# General comment: The approach described in this manuscript can provide a helpful expansion of the data analysis of lidar-ceilometers. How much information can be added to the data however depends on the input to the model. Here, the model is trained with observations representing a continental European aerosol. Thus the results are representative for this type of aerosol and regions/times where/when it occurs. The approach in general is able to add significant benefit to climatologies derived from lidar-ceilometer networks and should therefore be published. The presentation of methods and results is sound. Limitation of the applicability to specific situations have mostly clearly been addressed by the authors. The manuscript deals with as-far-as-possible exploitation of ceilometer data – that is good. But at the point of estimating mass concentrations (see below) I have concerns, because the results you show suggest that PM10 can be estimated within 10-20% accuracy by ceilometers, which I don’t think is generally true. Given the uncertainties and assumptions involved, the presented time series comparison may not even be representative for your sites at all times. Though inversion of optical data is often remarkably good-natured and your ‘calibration’ works for the related conditions and regional climate, this does not take into account the complexity of PM10 measurements which reflects in the +/- 25% measurement accuracy in the EC 2008/50 directive.

- We agree with the reviewer: the validation results obtained for volume and mass concentrations are not necessarily representative of other sites and times. We now clearly state this in the revised version (see specific modification to the text on this point reported below in the reply to general comment, point #2). In fact, it is the validation exercise at this specific site that found “that PM10 was estimated within 10-20% accuracy by ceilometer observations”, not the methodology proposed in this paper. In this respect, the paper clearly states that the model’s standard uncertainties in the retrieval of aerosol extinction and volume are within 30-40% (line 514). The retrieval of mass (PM) requires adding to this uncertainty the one related to particle density and signal quality. So, on a more general basis, we expect an uncertainty of the order of 50% when inferring aerosol mass from lidar measurements. Still, our validation exercise returned results well within this range.

#1 The manuscript presents a model-based approach to infer extinction coefficients, particle surface- and volume/mass-concentrations from backscatter coefficients measured by lidar-ceilometers, based on statistical relations. Mie-calculations are performed for empirical ranges of particle sizes and refractive indices, yielding conversion factors which are stored into a look-up table. As the influence of unspherical particles is shortly discussed, the Mie approach seems sufficient. The aerosol modal representation and refractive indices for model input are based on a comprehensive literature survey. Owing to the size distributions and the range of refractive indices used for the ensemble calculations, it is valid for ‘continental aerosol’. The inversion of ALC profiles uses state-of-the-art absolute calibration and the Rayleigh method according to Wiegner et al. 2012.

Evaluations are the most important part of the manuscript: The simulations (not including measurements) are evaluated against measurements: First the backscatter coefficient (BSC) vs Lidar ratio (LR) relation is compared against climatologies from EARLINET, CALIPSO and other networks. It is shown that average climatological LR are reproduced and that the frequency distribution of simulated BSC-LR pairs is roughly consistent with the corresponding distribution of EARLINET observations. There are, however, deviations, which are attributed to particle sizes and compositions which are, by design, not captured by the model.
Then, AOD inferred from CHM15k lidar-ceilometers are compared against each 1-2 years of data from 3 Italian stations with radiometers. Frequency distributions of the bias between inferred and measured AOD are shown and one example for illustration. The usual extrapolations of radiometers to 1064 nm and of the profile below 400m towards the ground (overlap) are done.

#2 Thirdly, volume and mass concentrations are estimated from lidar-ceilometers, based on the proposed model and compared to in-situ measurements with optical particle spectrometers. Given your limitation to ‘continental type aerosol’, the large variability in essential conversion factors showing up in the statistical evaluations, uncertainties due to the overlap extrapolation, I wonder how representative these results are. As OPS measure dry aerosol while lidar/ALC measure optical parameters under ambient conditions: I can hardly believe that the parametrisation in your model and the information about atmospheric humidity as such is accurate enough to allow proper humidity correction in the range of the uncertainties given here. I think that these results can only be achieved under very specific conditions - aerosol type, stratification, homogeneity etc. This should be discussed in more detail.

- We agree with many of the referee considerations here. As mentioned in the text (#lines 455-459), the comparison between OPC and ALC-retrieved volumes suffers from intrinsic factors where the different sampling conditions play a major role.

Regarding the RH impact, we report here our reply to reviewer #3 who also had a comment about the impact of RH on the ALC volume retrieval.

We know this effect is important in our volume estimates and relevant errors (e.g. also Barnaba et al., 2010; Adam et al., 2012). In fact, the RH dependence is taken into account by the model itself and it is accounted for in the model results variability. Indeed, errors can be much larger in the retrieval of PM loads, where a further unknown (particle density) is involved. In fact, we propose to retrieve volume not mass, and reference aerosol volume measurements are rather complex to perform if not including the full size distribution (as optical instruments do).

A further missing information would concern hygroscopicity of observed aerosols. We believe a full discussion (ALC vs other techniques) of volume comparisons would require a full paper itself. We believe it is better here to show some comparisons as in Figure 8, demonstrating the ALC volume estimates to well match the optical ones within the expected relevant variability.

However, to provide more information about RH, we added a horizontal bar in the upper part of Figure 8 indicating the range (RH<60%, 60%<RH<90% and RH>90%, respectively) of the measured in-situ RH during the ALC-OPC volume comparison.

The following text has been added:

This latter effect is confirmed by the large RH values (RH > 90%) measured after 18 UTC. The lower panel shows a good agreement between the ALC-derived and the Fidas OPC V_{a} values, in particular until 04 UTC and after 16 UTC. Some differences emerge around 07 UTC and between 11 and 15 UTC, where the ALC volume is lower by a factor of 2 compared to the in situ Fidas V_{a} values. The smaller minimum detectable size of the Fidas OPC instrument with respect to the OPS is likely the reason for the better accord between ALC and OPC V_{a} values in this test date. For this case, the effect of RH seems to be less important, and indeed RH values keep lower than 90%.

In general, high RH values (RH >= 90%) are known to markedly affect the aerosol mass estimation from remote sensing techniques and its relationship with ‘reference’ PM2.5 or PM10 measurements methods, usually performed in dried conditions (e. g Barnaba et al., 2010; Adam et al., 2012, Li et al., 2016, Li et al., 2017). This theme is also discussed in Diemoz et al. 2018a for the ALC measurement site of Figure'
Concerning the PM10 comparison, to specify the limitation of the obtained results, we added the following sentence in section 4.2.2:
This agreement attests that SPC site can indeed be considered an ‘average’ continental site and suggests the potential of this approach to derive information on aerosol volume and mass. Still, due to the specificity of each site and to the limited period considered here, these results cannot be taken as representative of all continental sites at all times. Further studies at different places and over longer time periods would be necessary to better assess the uncertainty of the proposed retrieval, including uncertainties due to the variability of ‘continental’ conditions (in terms of particle size distribution, compositions, hygroscopic effects, etc.), but also of the instrument-dependent performances (e.g. overlap corrections, etc.).

We also added the two following sentences to the manuscript conclusions:
1) ‘Overall, the good results obtained in our validation efforts are encouraging but necessarily related to the specific conditions at the sites and to the instrument characteristics considered. They are therefore not necessarily representative of results obtainable in all European continental sites, at all times. Further tests using wider datasets covering a variety of sites and ALC/lidar instrumentation would be desirable to better understand potential and limits of the applicability of the proposed method over the larger scale.’
2) Additionally, although our validation exercise returned results well within the uncertainties related to the model statistical variability alone (i.e., the relative errors associated to the mean functional relationships), the expected total uncertainty to be associated to the method should include terms that have not been specifically addressed in this work, as for example the instrumental error itself.

And modified the following sentence in the abstract:
‘Although limited in time, our comparison showed rather good agreement too. In particular, the ALC-derived daily-mean mass concentration for the considered site and specific period was found to well reproduce corresponding (EU regulated) PM10 values measured by the local Air Quality agency in terms of both temporal variability and absolute values’.

Specific comments:

#1 Line 80: : : and affordable for aerosol applications, : : :

- Done, thank you for this remark

#2 Line 108: this is at best true in a climatological sense, but not on shorter time scales. But even on the long-term, Putaud et al. 2010 report large differences in the aerosol distribution over Europe

- We reformulated the sentence in the following way:
‘we address here an ‘average-continental’ aerosol type (i.e. clean to moderately polluted continental aerosol conditions, e.g., section 2.1), expected to climatologically occur over most of Europe, despite the known differences that can be encountered across the continent both in the short and the long-term (e.g., Putaud et al. 2010).’

#3 Line 181: In this formula mRH converges to 2m0 for rmi_RH = rmi_0, i.e. for a large aequous droplet. Replace by mRh = mW + : : :

- Corrected, thank you for this remark
In eq. 4 and 5, rim_RH and miRH are the: should be: rmi_RH:

- Corrected, thank you for this remark

It is unclear to me what that means – what is 1%?

- We meant the $\beta_a$ region where each of the 10 equally-spaced bins per decade of $\beta_a$ contains at least 1% of the simulated points for the various aerosol parameters (i.e. $\alpha_a$, $S_a$ and $V_a$). We then reformulated the sentence:

The red vertical bars of Figure 2 also highlight the ranges of $\alpha_a$, $S_a$ and $V_a$ which are statistically significant, i.e. those in which, at $\lambda = 1064$ nm, the model provides at least 1% of the total points per corresponding bin of $\beta_a$.

- Corrected, thank you for this remark

But only as a statistical ensemble average over all data without evaluating the temporal correlation

- That is correct. We reformulated the sentence as follows:

Statistically, the highest number density of simulated data well fits the observations....

Hamburg (and the others as well) is not really a continental site but considerable sea-salt contribution can be expected in the coarse mode, (at least for Hamburg) likely not less on average than from dust. So does the statistical agreement with your model results confirm the significance of your model? Are you sure, you'd get a worse agreement e.g. for Mace Head and would you expect to be able to draw significant conclusions about the aerosol type?

- As presented in the text (#lines 320-322), for Hamburg, the distribution of LR values towards large values of $\beta_a$ (Fig. 4) could be due to the presence of sea-salt aerosols. This contribution does not appear using the relative LR difference (LRdif=0.05) at 355 nm as an indicator of the agreement between model and measurements. As suggested by the reviewer, to verify the statistical significance of LRdif, we computed the LRdif for the EARLINET station of Cork in Ireland (Mace Head was not available). In this case, the value of LRdif (=0.25) at 532 nm attests the presence of a significant difference between the model and measurements. This is correct because at this station the sea-salt contribution is predominant. Conversely, the Hamburg site (some 60 km from the sea) is mainly continental and affected by sea-salt aerosols mainly in summer and for a specific wind direction (Matthias and Bösenberg, 2002).

The LRdif values for these two stations show that we are able to draw some conclusions about the aerosol type, at least discriminating between mainly continental and non-continental sites. From our results, this was already clear for the Potenza and Lecce sites, whose results indicate that these sites were not purely continental (see appendix D).

why do you exclude desert dust days?

- This is because both optical and physical properties of desert dust are very different from those of ‘continental aerosol’ (and there is an important issue of aerosol non-sphericity in that
case, e.g., Barnaba and Gobbi 2001). We remind that, for the same reason, we also removed desert dust affected measurements from the EARLINET dataset when comparing modelled and measured LR in this work.

#10 Line 434: you should note that these conclusions are valid only in a statistical sense

- Done, thank you for this remark.

#11 Line 452: the data from 0-75m are those from 300 m a.g. (where overlap correction is feasible) extrapolated to the ground?

- No, for this system we didn't use the extrapolation to the ground but the original RCS data corrected by the O(z) function down to the lowermost atmospheric layers. In fact, as explained in section 4.2.1 #lines 394-395, the O(z) of ALC system at ASC is optimally characterized down to the ground.

#12 Line 444ff: You should specify that these OPC channel data are given as diameters, while above you mostly discuss sizes in terms of radii

- This has been specified, thank you.

#13 Line 480: what is the meaning of a particle density of 2 _g/m3? Typical densities are of the order 1000 kg/m3. There seems to be a conversion factor included.

- We apologize for this typo, we corrected to g/cm3.

#14 Line 486ff: Isn't the nocturnal boundary layer depth often in the range of few tens to hundreds of meters, even in summer? Strong vertical gradients in the lowest 200-300 m seem quite likely to me.

- In fact, our statistical (3-year) ALC record shows the mixing layer height at SPC to descend below 250 m only 4-5 hours per day in July (usually between 22 and 3 UTC, i.e., when emissions are at a minimum). We believe this contributes to the good agreement between the ALC and the PM10 measurements. We reformulated the sentence: The comparison to ground-level PM10 at SPC is expected to be only slightly affected by the height difference during the considered period of the year (i.e. June and July), particularly in daytime due to the strong convection in the boundary layer. Possible exception could be in nocturnal conditions when vertical gradients in the lowermost hundreds of meters can occur. However, our statistical (3-year) ALC records show the mixing layer height at SPC to descend below 250 m only 4-5 hours per day in July (usually between 22 and 3 UTC, i.e., when emissions are at a minimum).
General comments: The authors present an interesting study about retrieving aerosol properties (extinction coefficient (E), surface area (S) and volume (V)) from lidar and/or automated lidar-ceilometer (ALC) backscatter measurements. The key of the method is using a “Monte-Carlo” model to simulate the relationship between E, S, V and backscatter for different continental aerosol microphysical properties which could occur in real life and then implementing the relationships in the retrievals. Based on the 20000 model simulations, the relationship between lidar backscatter and aerosol E, S, V were investigated and dependence of lidar ratio (LR) to the backscatter at three lidar wavelengths (355 nm, 532 nm, 1064 nm) were fitted. The model-based LR were tested by comparing model simulations with raman lidar observations at 355 nm and found agree well with observations. Then the method was implemented to retrieve AOT and aerosol volume, PM10 and the results were compared with in-situ measurements. Although this method has some limitations in retrieving aerosol volume, mass, it shows the potential of using ALC for aerosol properties retrieval. The paper is well written and structured. The method was explained clearly, and main assumptions and limitations of the method were discussed. The topic is well suited for the AMT. I have a few comments and recommendations before the paper can be published.

We thank the reviewer for dedicating time to check and improve our manuscript. Following the constructive comments of the referee’s, several corrections have been made on the paper.

#1 Line 128: Both the step 1 and step 2 are about the aerosol model, why didn't authors put them both in the same section (section 2)?

- This choice was intended to separate methodology from results. Given the Reviewer’s objection we re-structured the text so that the old section 3 ('Model simulation results') is new section 2.2.

#2 Line 140: Is the r_mi at here same as the r_i in the equation 1 or it is another parameter? What are the m_(r i) and m_(im i)?

- We thank the referee for pointing out this inconsistency. All the notations indicated as misleading by the reviewer have now been corrected accordingly. In particular:
  - r_mi (indicating the modal radius) was replaced by ri as in eq.1
  - mr and mi (real and imaginary refractive indices, respectively) are now mr_i and mim_i.

#3 Line 143: what are the specific rules?

- The “specific rules” concern the ‘variability ranges for the number mixing ratio xi (Ni/Ntot) of each component to this total’. This latter definition replaces now the original one.

#4 Line 147: The description of m_(r i) and m_(im i) should be given at the first time when they appeared in the paper, see the related comment above. Secondly, more explanations about the real and imaginary refractive indices and how they are used in the aerosol optical properties calculation should be provided.
- The real and imaginary refractive indices are now introduced at this line (147), which is where these variables are used for the first time. Their usage in the calculation of the aerosol optical properties is specified later in the text (see equation 7-8). To clarify this, the following sentence has been added (lines 147-148): 'Being the result of different sources/processes, the three modes are also assumed to have a different composition, this impacting the optical computations through the relevant particle refractive index ($m_i$), with both its real and imaginary component ($m_i = m_{r,i} - i \times m_{im,i}$). The Mie theory for spherical particles of radius $r_i$ and refractive index $m_i$ are then used to compute the extinction and backscatter coefficients (see equations below).'

#5 Line 151: What is the exact size range? The authors should indicate the range or refer the tables which shows the range of the parameters at here. Same as the mode 1, 2 and 3.

- Thank you. We added the size ranges used for the three modes (Lines 151, 156, 160, respectively)

#6 Line 154: Why did the authors only use those values at 355 nm?

- In fact, the values at the three wavelengths addressed in the paper are provided in Table 2. In the text we only mentioned those at 355 nm as a quick reference. To clarify this, we now added the following sentence (line 154): 'A description of the assumptions made for each mode and relevant parameter, mostly based on literature data (Table 1), is given hereafter, the summary of the relevant variability chosen for each parameter being provided in Table 2.'

#7 Line 175-182: How did the authors decide to use those equations to stand the altitude dependence? Some references should be added here. What is the BG1? Is it the BG01 mentioned before?

- We added the relevant references (Patterson et al., 1980 and BG01). Yes, we erroneously used BG1 instead of BG01. This was corrected in the revised text.

#8 Line 197: The reference of the Mie theory or code should be added here.

- The reference to Bohren and Huffman (1983) has been added.

#9 Line 203: For mode 1 and mode 2, only the values of $m_{r,i}$ and $m_{im,i}$ at wavelength at 355 nm were introduced. How did the authors get the value at 1064 nm?

- Definition of the wavelength dependence of the refractive indexes employed for the different modes is specified at lines #164-168. Basically, we used the wavelength dependence reported in d’Almeida et al. (1991) and in Gasteiger et al. 2011, and Wagner et al., 2012.

#10 Line 215: The “(A)” should be after the “average”.

- Corrected, thanks.
#11 Line 242: The maxima is the maxima of the fitting curve but not the maxima of the all samples. Right?

-Correct. We added 'maxima of the fitting curve' in the sentence.

#12 Line 245: For the wavelength 1064 nm, there are some samples with LR larger than 80 based on the figure 3c.

- Right. We corrected the sentence in the following way: 'LR in the range 18 - 80 sr, except for a minor number of outliers'.

#13 Line 292: Are the relative errors the errors of lidar measurements? What are the standard measurements (truth)?

Yes, these are the errors associated to the EARLINET measurement used as reference in our study (e.g., Introduction)

#14 Line 298: 5 sites were chosen, but why there are only 4 sites depicted in the figure 4.

- This is because Figure 4 depicts the results of the model vs. measurements comparison in terms of LR vs $\beta_a$ at $\lambda=355$ nm. Unfortunately, the Madrid lidar system does not have the 355 nm emission wavelength. Still, we reported in Figure C1 (Appendix C) the corresponding results at $\lambda=532$ nm including Madrid (the Hamburg lidar system is missing in this case as it does not have the 532 nm emission wavelength). Following this comment, we now added the following sentence (line 299): 'For the EARLINET Raman stations fulfilling these requirements, Figure 4 depicts the results of the model-measurements comparison in terms of LR vs $\beta_a$ at $\lambda=355$ nm (the corresponding results at $\lambda=532$ nm, including Madrid in place of Hamburg, are given in Appendix C, Figure C1)'.

#15 Line 313: $\alpha'A^\dagger U\alpha'U_{mod,\alpha'A^\dagger U\alpha'A^\dagger}$ should be explained at here.

- The definition of LRmod and LRmeas has been added, (we guess this was the objection).

#16 Line 329: The table 6 should be referred at here.

- Thank you, the reference to Table 6 has been added (line 329).

#17 Line 366-371: Although the retrieval method was introduced by other scientists before, it is better to discuss more about how to derive aerosol extinction from the ALC e.g. show the key equations. Audiences may have questions like what are the raw data of the ALC? Are the raw data the range corrected backscatter? Does the raw data already consider the attenuation of signal from height z to surface due to aerosol and molecular extinction?

- The raw data of the ALC considered in this study is the range corrected signal $z^2P(z)$ where
z is the range, and (P) the raw signal stored in Netcdf format. To provide the information requested, we added the key equations of the algorithm at the end of section 3.2. Now the new text reads as follows:

“The forward solution of the Klett inversion algorithm is thus adopted here. In particular, to obtain \( \beta \) from ALC measurements, we report the equations of the detailed algorithm procedure described in Wiegner and Geiß (2012, equations 1–3):

\[
\beta(z) = \frac{Z(z)}{LR N(z)} - \beta_m(z) \tag{11}
\]

with

\[
Z(z) = LR z^2 P(z) \exp[-2 \int_0^z (LR \beta_m - \alpha_m) dz'] \tag{12}
\]

and

\[
N(z) = c_L - 2 \int_0^z Z(z') dz'. \tag{13}
\]

Here, \( \beta_m \) and \( \alpha_m \) are the molecular backscatter and extinction coefficients calculated from climatological, monthly air density profiles, and \( z^2 P(z) \) is the ALC range(z)-corrected-signal (P) (also referred to as RCS), that is the raw data format of the considered ALCs. Knowledge of the calibration constant \( c_L \) is needed to solve eqs. 13 (and thus 11, forward solution). When analyzing ALC daily records, the constant \( c_L \) has been obtained by the “backward approach” (Rayleigh calibration) applied to night-time, cloud-free ALC signal averaged over 1 or 2 hours at 75 m height resolution. This allows for using the best \( c_L \) retrieval (that is the night-time, lowest noise one), in the forward solution of the lidar equation, which guarantees operating over the best signal to noise range of the ALC signal.

#18 Line 389: What made the authors to choose this threshold of AOT for cases screen?

- Thank for noticing this omission. We now added the following sentence to explain this choice: ‘This range allows for excluding the data points with 1064nm AOT lower than the sunphotometer accuracy (dAOT=0.01) and those where we found aerosol extinction to cause significant deterioration in our ALC signal.

#19 Line 408-421: It is suggested to give a AOT VS AOT scatter plot at here. Then it will help the audiences to have sense of both absolute and relative errors of AOT.

- Following this suggestion, we added in Appendix E, the AOT vs AOT scatter plots for the three considered sites. The new text reads as follows:

To have sense of both absolute and relative errors of AOT, we reported in this section the scatter plots between the hourly-mean coincident AOTs at 1064 nm as derived by ALC model-based approach and those measured at 1020 nm by the sun-photometers installed at RTV, SPC and ASC, respectively (Figure E1, E2 and E3). The corresponding linear fit \( y = bx \) (red line), where \( x = \) sun-photometer AOT, \( y = \) Nimbus CHM15k AOT are also shown in the plots. The values of the correlation coefficients for the three sites (R = 0.77, R=0.72 and R=0.73 for RTV, SPC and ASC, respectively) attest a relatively good agreement between the two AOT
measurements.'
The three added scatter plots are shown hereafter:

Figure E1. Scatter plot between the hourly-mean coincident AOTs at 1064 nm as derived by the ALC model-based approach and measured at 1020 nm by the AERONET sunphotometer at RTV. The red line represents the linear fit $y = bx$ between the two datasets, where $x =$ sun-photometer AOT; $y =$ Nimbus CHM15k AOT.
Figure E2. Scatter plot between the hourly-mean coincident AOTs at 1064 nm as derived by the ALC model-based approach and measured at 1020 nm by the SKYRAD photometer at SPC. The red line represents the linear fit $y = bx$ between the two datasets, where $x =$ sun-photometer AOT; $y =$ Nimbus CHM15k AOT.
Figure E3. Scatter plot between the hourly-mean coincident AOTs at 1064 nm as derived by the ALC model-based approach and measured at 1020 nm by the SKYRAD photometer at ASC. The red line represents the linear fit $y = bx$ between the two datasets, where $x =$ sun-photometer AOT; $y =$ Nimbus CHM15k AOT.

#20 Line 431: With the fixed LR=52, the bias ($|dAOT| = 0.021$ and 0.006) are smaller than the model-based bias ($|dAOT| = 0.11$, 0.13) shown in line 428, right? authors said it is larger?

- Indeed it is larger. The problem was that the numbers reported in the text did not correspond to the values in Table 8. This was an error of us. Thank you for noting that. Actually, with fixed LR=52 sr, the bias at SPC and RTV is equal to 0.021 and 0.026, respectively, whereas the model-based bias is 0.011 and 0.013. The correct values have been inserted in the revised text.

#21 Line 440-444: How did the authors calculate the aerosol volume? Was the retrieval based on equation 7 and 10? Authors should explain more about the retrieval at here.

- We understand this point was not clear enough, we thus reformulated the relevant sentence as follows (line 441): ‘In particular, we use the model-estimated 7th-order polynomial fit equation linking $V_a$ and $f_a$ at $\lambda = 1064$ nm (see Table 3 and Figure 2c) to retrieve aerosol
volume profiles from ALC-derived βa measurements. These results were compared to ...

#22 Line 460-470: The hygroscopic growth could induce large differences between the insitu measured and the ALC retrieved aerosol volume. In the work of Siwei Li et al. (2016, 2017), they discussed the impacts of aerosol size distribution in the retrieval of PM2.5 using ceilometers (Li et al., 2016) and relationship between relative humidity and PM2.5/ceilometer-backscatter ratio (Li et al., 2017). More discussion about volume, PM retrieval and comparisons of model-based retrieval with in-situ measurements e.g. model vs in-situ scatter plot should be added here. Adding aerosol size (can compare the in-situ measurements and angstrom exponent) and relative humidity information and analysis at here may help the authors to support their conclusion.

- The referee is right and we know this effect is important in our volume estimates and relevant errors (e.g. also Barnaba et al., 2010; Adam et al., 2012). In fact, the RH dependence is taken into account by the model itself and it is accounted for in the model results variability. Indeed, errors can be much larger in the retrieval of PM loads, where a further unknown (particle density) is involved. In fact, we propose to retrieve volume not mass, and reference aerosol volume measurements are rather complex to perform if not including the full size distribution (as in the case of optical instruments).

A further missing information would concern hygroscopicity of observed aerosols. We believe an extensive discussion (ALC vs other techniques) of volume comparisons would require a full paper itself. We believe it is better here to show some comparisons as in Figure 8 demonstrating the ALC volume estimates can well match the optical ones within the expected relevant variability. However, to provide more information about RH, we added a horizontal bar in the upper part of Figure 8 indicating the range (RH<60%, 60%<RH<90% and RH>90%, respectively) of the measured in-situ RH during the ALC-OPC volume comparison.

In this respect, the following text has been added:

'This latter effect is confirmed by the large RH values (RH > 90%) measured after 18 UTC. The lower panel shows a good agreement between the ALC-derived and the Fidas OPC V_a values, in particular until 04 UTC and after 16 UTC. Some differences emerge around 07 UTC and between 11 and 15 UTC, where the ALC volume is lower by a factor of 2 compared to the in situ Fidas V_a values. The smaller minimum detectable size of the Fidas OPC instrument with respect to the OPS is likely the reason for the better accord between ALC and OPC V_a values in this test date. For this case, the effect of RH seems to be less important, and indeed RH values keep lower than 90%.

In general, high RH values (RH >= 90%) are known to markedly affect the aerosol mass estimation from remote sensing techniques and its relationship with ‘reference’ PM2.5 or PM10 measurements methods, usually performed in dried conditions (e. g. Barnaba et al., 2010; Adam et al., 2012, Li et al., 2016, Li et al., 2017). This theme is partially discussed in Diemoz et al. 2018a for the ALC measurement site of Figure 8.'

#23 Line 477: What specific aerosol densities did the authors use in the retrieval and why?

- Actually, values of aerosol densities were already mentioned in the text (#line 480: ρ_a = 2 g/cm^3, with a range between 1.5-2.5 g/cm^3). We took the opportunity of this comment to specify that: 'This range covers approximately the mean ρ_a values of the SPC site.'

#24 Line 485: Why did the authors use different heights in estimation of surface aerosol volume (0-75 m) and mass (at 225 m)?
- This is because the two estimates come from different systems. As explained at #lines 394-395, the ALC overlap function of the ASC site has been optimally characterized and therefore for this system we used the lowest altitudes to estimate the surface aerosol volume. Conversely, the ALC system at SPC has an old firmware and its overlap function is not optimally characterized, we therefore used the 225 m level as more trustworthy. This was highlighted in line 485.

#25 Line 488-490: Were the mean and relative difference between the two-series based on hourly average PM10 or daily average PM10? What is the absolute difference? What is the R between them?

- As reported at #lines 474 and 481, the two series are the daily average PM10. We added in the text the values of R and of the absolute and relative differences (line 488): ‘Overall, Figure 9 confirms a good agreement between the ALC-derived and the ARPA reference PM10 values, with a correlation coefficient (R) of 0.73. In fact, mean, absolute mean and relative differences, between the two series are: \(<dPM10> = 2.8 \pm 6.5 \text{ g/cm}^3, \langle|dPM10|\rangle = 5.2 \pm 4.7 \text{ g/cm}^3 \text{ and } \langle(dPM10/PM10)\rangle = 0.15 \pm 0.27.'

#26 Line 496: Where is the close bracket?

- It was missing, sorry. Corrected.

#27 Line 549: Surface area and volume are not the optical properties. The results from this work showed that ancillary data information are needed to get accurate aerosol properties e.g. volume, mass.

- The sentence has been reformulated in the following way: ‘On the other hand, the proposed approach has the main advantage of allowing the operational (i.e. 24/7) retrieval of fairly reliable, remote sensing profiles of aerosol optical (\(\alpha_a, \beta_a\)) and physical (\(S_a, V_a\)) properties (with associated uncertainties and limitations) by means of relatively simple and robust instruments.

#28 Line 551: What kind of meteorological monitoring can be provided by the method?

- Potentially, the aerosol vertical characterization in terms of aerosol backscatter, extinction, surface and volume derived by the proposed-method together with the ALC ‘standard’ information on cloud base and on the boundary layer can provide interesting information on the aerosol-cloud interaction and the involved meteorological processes. We have integrated the sentence: ‘This could temporally and spatially complement the information coming from more advanced lidar networks (for example, the Raman channel of multi-wavelength system cannot be used in daylight conditions) and, more in general, could represent a valid option to deliver, in quasi real time, the 3D aerosol fields useful for operational air quality (e.g. integration of the in situ surface measurements) and for meteorological and climate monitoring (e.g. aerosol-cloud interaction and aerosol transport and dispersion processes).
A multi-wavelength numerical model in support to quantitative retrievals of aerosol properties from automated-lidar-ceilometers and test applications for AOT and PM10 estimation

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Abstract. Knowledge of the vertical distribution of aerosol particles is a key factor in the study of climate, air pollution, and meteorological processes. The use of automated lidar-ceilometers (ALCs) systems for the aerosol vertically-resolved characterization has increased in the recent years thanks to their low construction and operation costs, and to their capability at providing continuous, unattended measurements. At the same time there is a need to convert the ALC signals into usable geophysical quantities. In fact, the quantitative assessment of the aerosol properties from ALC measurements and the relevant assimilation in meteorological forecast models is amongst the main objectives of the EU COST Action TOPROF (Towards Operational ground-based PROFiling with ALCs, doppler lidars and microwave radiometers). Concurrently, the E-PROFILE program of the European Meteorological Services Network (EUMETNET) focuses on the harmonization of ALC measurements and data provision across Europe. Within these frameworks, we implemented a model-assisted methodology to retrieve key aerosol properties (extinction coefficient, surface area and volume) from elastic lidar and/or ALC measurements. The method is based on results from a large set of aerosol scattering simulations (Mie-theory) performed at UV, visible and near IR wavelengths using a "Monte-Carlo" approach to select the input aerosol microphysical properties. An average "continental aerosol type" (i.e. clean-to-moderately polluted continental aerosol conditions) is addressed in this study. Based on the simulation results, we derive mean functional relationships linking the aerosol backscatter coefficients to the above-mentioned variables. Applied in the data inversion of single wavelength lidars and/or ALCs, these relationships allow quantitative determination of the vertically-resolved aerosols backscatter, extinction, volume and surface area and, in turn, of the extinction-to-backscatter ratio (i.e., the lidar-ratio, LR) and of extinction-to-volume conversion factor (c_v) at 355, 532, 1064 nm. These variables provide valuable information for visibility, radiative transfer and air quality applications. This study also includes 1) validation of the model simulations with real measurements and 2) test applications of the proposed model-based ALC inversion methodology. In particular, our model simulations were compared to backscatter and extinction coefficients independently retrieved by Raman lidar systems operating at different continental sites within the European Aerosol Research Lidar NETwork (EARLINET). This comparison shows good model-measurements agreement, with LR discrepancies below 20%. The model-assisted quantitative retrieval of both aerosol extinction and volume was then tested using raw data from three different ALCs systems (CHM15k-Nimbus), operating within the Italian Automated Lidar-ceilometer Network (ALICENET). To this purpose, a one-year-record of the ALCs-derived aerosol optical thickness (AOT) at each site was compared to direct AOT measurements performed by co-located sun-sky photometers. This comparison shows an overall AOT agreement within 30% at all sites. At one site, the model-assisted ALC estimation of the aerosol volume and mass (i.e., PM10) in the lowermost levels was compared to values measured at the surface-level by co-located in situ instrumentation. Within this exercise, the ALC-derived daily-mean
mass concentration was found to well reproduce the corresponding (EU regulated) PM10 values measured by the local
Air Quality agency in terms of both temporal variability and absolute values. Although limited in space and time, the
good performances of the proposed approach in these preliminary tests suggest it could possibly represent a valid option
to extend the capabilities of ALCs at providing quantitative information for operational air quality and meteorological
monitoring.

1 Introduction

Due to the impact of atmospheric aerosols on both air quality and climate, substantial efforts have been made to expand
our knowledge of their sources, properties and fate. Aerosol particles affect the Earth’s radiation budget mainly by two
different processes: 1) by scattering and absorbing both solar and terrestrial radiation (aerosol direct effect, Haywood
and Boucher, 2000 and aerosol semi-direct effect, Johnson et al., 2004) and 2) by serving as cloud and ice condensation
nuclei (aerosol indirect effect, Loehmann and Feichter, 2005, Stevens and Feingold, 2009 and Feingold et al., 2016). The
complexity of these processes and the extreme spatial and temporal variability of the aerosol sources, physical and
chemical properties and atmospheric processing make the quantification of their impacts very difficult. Aerosols have
also proven detrimental effects on human health (e.g., D’Amato et al., 2013, World Health Organization, 2013,
Lelieveld et al., 2015). In fact, their concentration (often evaluated in terms of particulate matter mass, or PM) is
regulated by specific air quality legislation worldwide. In Europe, the Air Quality Directive 2008/50 defines the
‘objectives for ambient air quality designed to avoid, prevent or reduce harmful effects on human health and the
environment as a whole’ (EC, 2008).

Among the aerosol observational systems, the LIDAR technique has been proved to be the optimal tool to provide
range-resolved, accurate aerosol data necessary in radiative transfer computations (e.g. Koetz et al., 2006, Tosca et al.,
2017) and is often usefully employed in supporting air quality studies (e.g. Menut et al., 1997, He et al., 2012). With a
spectrum of different system types (elastic backscatter, Raman, High Spectral Resolution, and multi-wavelength lidars),
each with specific pro and cons (Lolli et al., 2018), this technique allows retrievals of aerosol and cloud optical
properties and relevant distribution within the atmospheric column at several ground-based observational sites (Fernald
et al., 1972; Klett, 1981; Shipley et al., 1983, Kovalev and Eichinger, 2004, Heese and Wiegher, 2008; Ansmann et al.,
2012). Since 2006, the Cloud Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) platform (Winker
et al. 2003) also provides a unique, global view of aerosol and cloud vertical distributions through space-based
observations (at the operating wavelengths of 532 and 1064 nm). Recently, within the Cloud-Aerosol Transport System
(CATS) mission, a lidar was also installed at the International Space Station (ISS, McGill et al., 2015 and York et al.,
2016). Space-borne lidar observations are however affected by some drawbacks, as: 1) limited temporal resolution and
spatial coverage (the CALIPSO spatial distance between two consecutive ground tracks is about 1000 kilometers and
each track has a footprint of 70 m), 2) the contamination of unscreened clouds, and 3) difficulties in quantitatively
characterizing the aerosol properties in the lowermost troposphere (Pappalardo et al., 2010). Ground-based lidar
networks thus still represent key tools in integrating space-borne observations to study aerosol properties and their 4D
distribution. An example of these networks is the European Aerosol Research Lidar NETwork (EARLINET,
http://www.eearlinet.org/), which, since 2000, provides an extensive collection of ground-based data for the aerosol
vertical distribution over Europe (Bösenberg et al., 2003, Pappalardo et al., 2014). The advanced multi-wavelength
elastic and Raman lidars employed in this network allows independent retrieval of aerosol extinction ($\alpha_a$) and
backscattering coefficient ($\beta_a$) profiles. Yet, despite their unsurpassed potential in data accuracy, advanced lidar
networks such as EARLINET have the unsolved problems of the sparse spatial and temporal sampling and of the
complexity of operations. In fact, the typical distance between the EARLINET stations is of the order of several hundreds of kilometers and regular measurements of EARLINET are only performed on selected days of the week (Mondays and Thursdays) and for a few hours (mainly at nighttime, due to low signal-to-noise ratio of the Raman signal in daylight). Furthermore, these systems are complicated to be operated, require specific expertise and are therefore unsuitable for operational applications.

Nowadays, hundreds of single channel Automated Lidar Ceilometers (ALCs) are in operation over Europe and worldwide. Although such simple lidar-type instruments were originally designed for cloud base detection only, the recent technological advancements make now these systems reliable and affordable for aerosol measurements, increasing the interest in using this technology in different aerosol-related sectors (e.g. air quality, aviation security, meteorology, etc.). Recent studies showed that the ALC technology is now mature enough to be used for a quantitative evaluation of the aerosol physical properties in the lower atmosphere (Wiegner, M. and A. Geiß, 2012, Wiegner et al., 2014) and the exploitation of the full potential of ALCs in the aerosol remote sensing is a current matter of discussion in the lidar community (e.g. Madonna et al., 2015, 2018). The evaluation of ALC capabilities at providing quantitative aerosol information is among the main objectives of the EU COST Action ES1303, TOPROF (Towards Operational ground-based PROFiling with ALCs, doppler lidars and microwave radiometers). An effort in this direction is also underway in the framework of E-PROFILE, one of the observation programs of the EUropean METorological services NETwork (EUMETNET). In fact, several ALC stations are progressively joining E-PROFILE to develop an operational network to produce and exchange ALC-derived profiles of attenuated backscatter. A recent project funded by the EU LIFE+ program (DIAPASON, Desert-dust Impact on Air quality through model-Predictions and Advanced Sensors ObservatioNs, LIFE+:2010 ENV/IT/391) also prototyped and tested an ALC system with an additional depolarization channel, capable of discriminating non spherical aerosol types, such as desert dust (Gobbi et al., 2018). Such upgraded ALC systems could further improve the capabilities of the operational aerosol profiling in a near future.

Given the necessity to couple advancement in instrumental technology with tools capable of translating raw data into a robust, quantitative and usable information, we propose and characterize here a methodology to be applied to elastic backscatter lidars and/or ALC measurements to retrieve, in a quasi-automatic way, vertically-resolved profiles of some key aerosol optical and microphysical properties. This effort is intended to contribute better exploit these systems potential in integrating data collected by more advanced lidar systems/networks. In particular, the ALC-derived aerosol properties addressed in this study are: backscatter ($\beta_a$, $\text{km}^{-1} \text{sr}^{-1}$), extinction ($\alpha_a$, $\text{km}^{-1}$), surface area ($S_a$, $\text{cm}^2\text{cm}^{-3}$) and volume ($V_a$, $\text{cm}^3\text{cm}^{-3}$), the latter being convertable into aerosol mass concentration ($\mu \text{g m}^{-3}$) via assumption on particle density. To this purpose, we developed a numerical aerosol model to perform a large set of aerosol scattering simulations. Based on results from this numerical model, we derive mean functional relationships linking $\beta_a$ to $\alpha_a$, $S_a$ and $V_a$ respectively. These relationships are then applied in the ALC data inversion and analysis. A similar approach was applied in past studies for lidar-based investigations of stratospheric (Gobbi, 1995) and tropospheric aerosols (maritime, desert dust and continental type) at visible and UV lidar wavelengths, (Barnaba and Gobbi, 2001, Barnaba and Gobbi, 2004a, hereafter BG01, BG04a, respectively, Barnaba et al., 2004). Here we extend this approach to all the Nd:YAG laser harmonics commonly used by both advanced lidars and ALC systems (i.e. 355, 532, 1064 nm wavelengths) and specifically address an "average-continental" aerosol type, intended to represent clean-to-moderately polluted continental aerosol conditions (see Section 2.1). In fact, despite the known differences that can be encountered across the continent both in the short and the long-term (e.g., Putaud et al., 2010), this aerosol type is expected to climatologically dominate over most of Europe.
Overall, this investigation is organized as follows: in Section 2 we describe the aerosol model set up to reproduce clean to moderately polluted continental conditions, and the Monte Carlo methodology followed to compute the corresponding bulk optical and physical properties. Section 3 shows and discusses the results of the numerical model and presents the model-based, mean functional relationships linking the different variables at 355, 532 and 1064 nm. In Section 4 we evaluate both the model simulations capability to reproduce real measurements in continental aerosol conditions, and the capability of the model-based ALC inversion approach to derive quantitative geophysical information. The EARLINET database was used for the first task while tests on the accuracy of the model-based ALC inversion were performed evaluating both the ALC-derived aerosol volume and optical thickness (AOT, i.e. the vertically integrated aerosol extinction). To this purpose we applied the proposed methodology to three ALC systems operating within the Italian Automated LIdar-CEilometer NETwork (ALICE-NET, www.alice-net.eu). In particular, the ALC-derived AOT and aerosol volume (plus mass) were compared, respectively, to reference measurements performed by ground-based sun photometers and in situ aerosol instruments (optical counters and PM10 samplers).

Section 5 summarizes the developed approach and main results, critically examining strengths and weaknesses. It also includes discussion on the perspectives of the application of this (or similar) methodology in operational ALC networks.

2 The aerosol model

A numerical aerosol model was set up to calculate mean functional relationships between the aerosol backscatter ($\beta_a$) and some relevant aerosol properties, as $\alpha_a$, $S_a$ and $V_a$. This is done in a two-step procedure (Figure 1), following an approach similar to that developed by BG01 and BG04a:

1) Generation of a large set (here 20000) of aerosol optical and physical properties by randomly varying, within appropriate ranges, the microphysical parameters describing the aerosol size distribution and composition (blue box in Fig. 1);

2) Based on results at point 1), determination of mean functional relationships linking such key variables (grey box in Fig. 1).

The following Section describes rationale and set-up of the first step, the second one being thoroughly discussed in Section 3.

2.1 Selection of the aerosol microphysical parameters

As anticipated, an average ‘continental’ aerosol type (i.e. describing clean to moderately polluted continental conditions, e.g. Hess et al., 1998) was targeted in this study, this being the aerosol type expected to dominate over Europe. Based on a scheme originally proposed by d’Almeida et al. (1991), and on a large set of following observational evidences (e.g. Van Dingenen et al., 2004), in this work its size distribution is described as an external mixture of three size modes. These are (in order of increasing size range): 1) a first ultrafine mode; 2) a second fine mode, mainly composed of water-soluble particles; 3) a third mode of coarse particles.

A three-mode lognormal size distribution described by Eq. (1) is employed to this purpose:

$$\frac{dN}{d \log r} = \sum_{i=1}^{3} \frac{N_i}{\sqrt{2\pi} \log \sigma_i} \exp \left\{ -\frac{(\log r - \log r_i)^2}{2(\log \sigma_i)^2} \right\}$$

(1)
In Eq. (1), \( r_i \), \( \sigma_i \) and \( N_i \) are respectively the modal radius, the width and the particle number density of the \( i \)th aerosol mode (\( i = 1, 2, 3 \)). At each computation, \( r_i \) and \( \sigma_i \) are randomly chosen within a relevant variability range. Values of \( N_i \) are conversely obtained by firstly randomly choosing the total number of particles, \( N_{tot} \), to be included in the whole size distribution (\( N_{tot} = N_1 + N_2 + N_3 \)), and then by applying specific rules for the number mixing ratio, \( x_i = N_i/N_{tot} \), of each component to this total. To reproduce clean to moderately polluted continental conditions, the value of \( N_{tot} \) is made variable between \( 10^4 \) and \( 3 \times 10^4 \) cm\(^{-3} \) (e.g. Hess et al., 1998; Van Dingenen et al., 2004). Being the result of different sources/processes, the three modes are also assumed to have a different composition, this impacting the optical computations through the relevant particle refractive index (\( m_i \)), with both its real and imaginary component (\( m_i = m_r + i \times m_im \)). The Mie theory for spherical particles of radius \( r_i \) and refractive index \( m_i \) is then used to compute the extinction and backscatter coefficients (see below).

A description of the assumptions made for each mode and relevant parameter, mostly based on literature data (Table 1), is given hereafter, the summary of the relevant variability chosen for each parameter being provided in Table 2.

1) First Mode

This ultrafine mode is the one more directly simulating fresh, anthropogenic emissions. The number mixing ratio \( x_{i=1} \) (\( N_{i=1}/N_{tot} \)) of this mode is let variable between 10% (rural conditions, Van Dingenen et al., 2004) and 60% (more polluted conditions, Hess et al., 1998). The variability of its modal radius (\( r_{i=1} = 0.005 - 0.03 \mu m \)) is chosen to include from nucleation mode particles to Aitken mode particles. To take into account the wide variability of species within this ultrafine mode, from non-absorbing (e.g., inorganic particles) to highly absorbing materials (e.g. black carbon), wide ranges of variability have been set for its refractive indexes (at \( \lambda = 355 \) nm: \( m_{im,1} \) in the range 1.40 - 1.8, and \( m_{im,3} \) in the range 0.01 – 0.47, see Table 2 for the corresponding values at \( \lambda = 532 \) and 1064 nm).

2) Second Mode

The second aerosol mode accounts for 40-90% of \( N_{tot} \), with (dry) \( r_i \) between 0.03 and 0.1 \mu m. Its composition (\( m_{r,2} \), and \( m_{im,2} \)) is also made highly variable so to include water soluble inorganic and organic particles (Hess et al., 1998; BG04a; Dinar et al. 2008). In this case, at \( \lambda = 355 \) nm, \( m_{r,2} \) is in the range 1.40 - 1.7 and \( m_{im,2} \) is in the range 0.0001 – 0.01 (Table 2).

3) Third mode

This coarser aerosol mode (modal radius \( r_3 \) in the range 0.3 – 0.5 \mu m) is mainly intended to account for soil derived (dust-like) particles that are a primary continental emission. A quite narrow variability is thus fixed for its \( m_{r,3} \) and \( m_{im,3} \) values (1.5 – 1.6 and 0.0001 – 0.01, respectively at 355 nm). The relevant number mixing ratio \( x_3 \) (\( N_3/N_{tot} \)) is set variable between 0.001% and 0.5 %, this mode contributing mostly to the total aerosol volume (thus mass) rather than to the total number of particles.

As mentioned, refractive indexes were also made wavelength dependent, as this feature is also typically observed as linked to the different particle composition. In particular, for the second mode (water-soluble particles) we include an increase with the wavelength of the upper boundary values of \( m_{im,2} \) and a decrease of \( m_{r,2} \) at \( \lambda = 1064 \) nm (d’Almeida et al., 1991). For the (dust-like) third-mode particles, the upper boundary values of \( m_{im,3} \) are set to decrease with increasing wavelengths (Gasteiger et al., 2011, Wagner et al., 2012).

For convenience, the aerosol parameters boundaries summarized in Table 2 refer to dry particles and to ground level. However, the effect of a variable RH, its variability with altitude as well as the generally observed decrease of particle
number with altitude is also considered in the model. More specifically, the number of particles in each mode, $N_i$, and RH are both made altitude-dependent through the following equations (Patterson et al., 1980, BG01):

$$N_i(z) = N_i(0) \times \exp \left( \frac{-z}{H_i} \right),$$  

(2)

$$RH(z) = 70 \times \exp \left( \frac{-z}{5.5 \text{ km}} \right) \times (1 + dRH),$$  

(3)

the altitude $z$ being variable here between 0 and 5 km. $N_i(0)$ and $H_i$ in eq.2 are the number of particles at the ground and the scale height for each mode, respectively.

To describe the altitude effect, in eq. (2) an exponential decrease with height of the particle number density is assumed. To rescale the particle number density of the different modes, $H_{i=1-2}$ is set equal to 5.5 km (Barnaba et al., 2007) while $H_{i=3}$ (coarse particles) is set to 0.8 km (Barnaba et al., 2007). In eq. (3), the additional term $(1+dRH)$ allows a further variability with respect to the mean RH(z) profile assumed, here $dRH$ is randomly chosen between -60 and +60). Values of RH greater than 95% are discarded to avoid divergence.

Additionally, while first and third modes are assumed to be water insoluble, the second mode ($i=2$) is fully hygroscopic. Aerosol humidification is thus considered to act on both particle size and refractive indices of the second aerosol mode (e.g., BG01), as:

$$r_{2,RH} = r_{2,0} \frac{1 + 0.01RH}{2(1 + 0.01RH)},$$  

(4)

$$m_{2,RH} = m_w + (m_{2,0} - m_w) \left( \frac{r_{2,0}}{r_{2,RH}} \right)^3,$$  

(5)

In eq. 4 and 5, $r_{2,RH}$ and $m_{2,RH}$ are the RH-corrected modal radius and refractive index for the second mode, respectively; $r_{2,0}$ and $m_{2,0}$ are the particle dry modal radius and refractive index, respectively; $m_w$ is the water refractive index (assumed as equal to 1.34 – i7e°, 1.33 - i1.3e° 1.33 - i2.9e° at 355, 532 and 1064 nm, respectively).

Finally, following Barnaba et al. (2007), an increase of the width of the size distribution with altitude (eq. 6) has been introduced for the first and second aerosol mode:

$$\sigma_{1,2}(z) = \sigma_{1,2,0} \times \exp \left( \frac{z}{300} \right),$$  

(6)

In fact, Barnaba et al., (2007) showed that this was necessary to better reproduce the observed decrease of the Lidar Ratio (LR) with altitude, and likely related to a broadening of the particle size distribution with aging.

Once the value of each microphysical parameter is randomly selected within its relevant variability range, and once corrections are applied following eqs. (2) – (6), each resulting aerosol size and composition-resolved distribution is used to compute the aerosol $S_a$ and $V_a$, as well as to feed a Mie code (assumption of spherical particles, Bohren and Huffman, 1983) to compute $\beta_a$ and $\alpha_a$ (BG01, see also Fig. 1). Overall, the equations used are as follows:

$$\beta_a = \int Q_{bsc}(r, \lambda, m) \frac{1}{\ln 10} \frac{dN}{d\log r} \frac{dr}{r},$$  

(7)

$$\alpha_a = \int Q_{ext}(r, \lambda, m) \frac{1}{\ln 10} \frac{dN}{d\log r} \frac{dr}{r},$$  

(8)

$$S_a = 4\pi \int r^2 \frac{1}{\ln 10} \frac{dN}{d\log r} \frac{dr}{r},$$  

(9)
\[ V_a = \frac{4}{3} \pi \int r^2 \frac{dN}{d\log r} \frac{1}{r} dr, \quad (10) \]

where \( Q_{\text{bsc}}(r, \lambda, m) \) and \( Q_{\text{ext}}(r, \lambda, m) \) are, respectively, the backscatter and the extinction efficiencies. As mentioned, the optical computations are made at the three different wavelengths: 355, 532, 1064 nm (i.e., those of Nd:YAG laser harmonics, the most common wavelengths used by ground-based and space-borne aerosol lidars).

Since in our simulations the third aerosol mode is intended to represent dust-like particles, an empirical correction for non-sphericity is also applied to the Mie-derived optical properties of this mode. This procedure is based on BG01, which uses the results of Mishchenko et al. (1997) obtained for surface-equivalent mixtures of prolate and oblate spheroids.

### 2.2 Model simulation results

In Figure 2 we show the results of 20000 simulations of continental aerosol optical and physical properties derived randomly varying the relevant aerosol size distributions and compositions as described in the previous section. In particular, the results for \( \alpha_a, S_a \), and \( V_a \) are shown as a function of \( \beta_a \) in Figure 2a, b, c (blue crosses) referring to \( \lambda = 1064 \text{ nm} \). For each variable (A), average value per bin of \( \beta_a \) and relevant standard deviations \( (A) \pm dA \) are shown as red dots and vertical bars, respectively. Note that 10 equally spaced bins per decade of \( \beta \) have been considered, and that \( (A) \pm dA \) are only shown for bins containing at least 1\% of the total number of pairs. Corresponding relative errors \( (dA/\langle A \rangle) \) are depicted in Figure 2d, e, f. Some sensitivity tests of these model outputs to the variability of the input microphysical parameters employed are provided in Appendix A.

Based on these results, at step-two of the procedure (see scheme in Figure 1), we derive aerosol-specific mean relationships linking aerosol extinction, surface area and volume \( (\alpha_a, S_a \text{ and } V_a) \) to its backscatter \( (\beta_a) \). To this purpose, we used a seventh-order polynomial fit in log-log coordinates. The choice of a seventh-order polynomial fit was made for homogeneity with BG01 and BG04a. These relationships are shown as green lines in Figure 2a, b, c while the relevant fit parameters are reported in Table 3 referring to \( \lambda = 1064 \text{ nm} \) (fit parameters related to computations at \( \lambda = 355 \text{ and } 532 \text{ nm} \), are given in Table A1 and Table A2, Appendix B).

The red vertical bars of Figure 2 also highlight the ranges of \( \alpha_a, S_a \text{ and } V_a \), which are statistically significant, i.e. those in which, at \( \lambda = 1064 \text{ nm} \), the model provides at least 1\% of the total points per corresponding bin of \( \beta_a \). These are: \( 10^{-4} - 10^{-1} \text{ km}^2/\text{cm}^2 \), \( 10^{-7} - 10^{-5} \text{ cm}^2/\text{cm}^3 \) and \( 10^{-13} - 10^{-10} \text{ cm}^3/\text{cm}^3 \), for \( \alpha_a, S_a \text{ and } V_a \) respectively, corresponding to the backscatter range \( 9 \times 10^{-5} \leq \beta_a \leq 4 \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1} \). In terms of aerosol properties variability, the relative errors associated to \( \alpha_a \) and \( V_a \) show almost no dependence on \( \beta_a \), with values between 30\% and 40\%. Conversely, the modeled aerosol surface area exhibits a larger dispersion, with relative error values spanning the range 40\% - 70\%, and decreasing as \( \beta_a \) increases.

A key parameter for the inversion of lidar signals is the so-called Lidar Ratio (LR), i.e. the ratio between \( \alpha_a \) and \( \beta_a \) (Ansmann et al., 1992). In Figure 3 we thus show the results of our simulations in terms of LR vs \( \beta_a \) at the three \( \lambda \) (355, 532 and 1064 nm, Figure 3a, b, c, respectively) and relevant dLR/LR values (Figure 3d, e, f, respectively). The color code is the same of Fig. 2. Additional horizontal black lines have been inserted representing values (solid central lines) of the `weighted-LR' \( \pm 1 \) standard deviation (dotted side lines), i.e. the LR weighted by the number of simulated points in each considered backscatter bin. The `weighted-LR' values derived at 355, 532 and 1064 nm, are \( 50.1 \pm 17.9 \text{ sr} \), \( 49.6 \pm 16.0 \text{ sr} \) and \( 37.7 \pm 12.6 \text{ sr} \), respectively. Figure 3 also allows showing that the statistically significant regions of simulated backscatter values shifts towards smaller values with increasing \( \lambda \) (e.g. at \( \lambda = 355 \), the \( \beta_a \) extending regions is \( 4 \times 10^{-3} - 2 \times 10^{-2} \text{ km}^{-1} \text{ sr}^{-1} \), whereas, at 532 nm, it ranges between \( 2 \times 10^{-3} - 1 \times 10^{-2} \text{ km}^{-1} \text{ sr}^{-1} \)). Furthermore, Figure 3...
reveals a quite different shape of the LR vs $\beta_a$ functional relationships (green curves) at different wavelengths. At 355 and 532 nm the curve is concave, with quite similar LR maxima of the fitting curve (54.3 and 53.8 sr at approximately $\beta_a = 4 \times 10^4$ km$^{-1}$ sr$^{-1}$ and $2 \times 10^3$ km$^{-1}$ sr$^{-1}$, respectively). At 1064 nm the curve is conversely monotonic, with a flex point at $\beta_a = 3.4 \times 10^4$ km$^{-1}$ sr$^{-1}$. A larger data dispersion also characterizes the results at $\lambda = 355$ and 532 nm (LR values from 10 to 90 sr) in comparison to $\lambda = 1064$ nm (LR in the range 18 – 80 sr, except for a minor number of outliers). This translates into different LR relative errors at UV, VIS and infrared (IR) wavelengths. At 1064, dLR/LR slightly decreases for increasing backscatter, with values around 35%. At the shorter wavelengths, it increases as a function of $\beta_a$ with a large (>40%) relative error for values of $\beta_a > 2 \times 10^3$ km$^{-1}$ sr$^{-1}$.

To insert our results into a more general context, we compared the derived, model-based weighted-LR values to some LR data reported in the literature (Table 4). In particular, we selected some of the works using the aerosol model developed to invert the Calipso lidar data (Omar et al., 2009). This latter considers six different aerosol sub-types: clean continental (CC), clean marine (CM), dust (D), polluted continental (PC), polluted dust (PD), and smoke (S). Our model-derived LR at 532 nm falls in the middle of the range (35-70 sr) fixed by the Calipso CC and PC aerosol classes. The work by Papaggianopoulos et al. (2016), in which the LR values are adjusted accordingly to EARLINET observations, reports a LR range at 532 nm of 47-62 sr. At the same wavelength, the aerosol range defined by the LIVAS climatology (LIdar climatology of Vertical Aerosol Structure for space-based lidar simulation studies, Amiridis et al., 2015), is 54-64 sr. In both cases, our model seems to be closer to the LR values of CC aerosol type, which is compatible to our intention to simulate clean-to-moderately polluted continental aerosol type. At 532 nm, our LR value is also reasonably in between the CC and PC LR values derived by Omar et al. (2009), but again closer to the CC LR value. The very small decrease of LR values between 532 and 355 nm estimated by LIVAS for the CC aerosol is also consistent with our results. Similarly, our model predicts a lower mean LR in the near IR with respect to the green, in agreement with results of Amiridis et al. (2015) in CC conditions and not to those in polluted conditions. Table 4 also includes the continental aerosol LR values estimated in the work of Düsing et al. (2018) through comparison between airborne in situ and ground-based lidar measurements. Our model is in good agreement with their LR values at 355 and 532 nm. At 1064 nm, the algorithm developed by Düsing et al. (2018) provided a value of LR around 15 sr. On the other hand, in the same study the authors found that, rather, a value of LR = 30 sr gives the better accord between their Mie and lidar-based $\alpha_a$, this value being closer to our model-derived one at 1064 nm (LR = 37.7). The difference between these two values is explained by the authors to be probably due to the estimation of the aerosol particle number size distribution, a critical parameter for a reliable modeling of aerosol particle backscattering.

As a last added value of the outcome from our model-based results, we derive here and provide in Table 5 extinction-to-volume conversion factors, $c_v = V/\alpha_a$ (e.g., Ansmann et al., 2010) at three different wavelengths (355, 532 1064 nm), and compare these to similar outcomes from other studies. To our knowledge, values of continental particles $c_v$ at these wavelengths are only available in Mamouri and Ansmann (2017). Note that $c_v$, is also proportional, through the particle density $\rho_a$, to the inverse of the so-called "mass-to-extinctions efficiency" (MEE, i.e. $\alpha_a(V_c \rho_a)$) a parameter important in several aerosol-related applications (e.g. the estimation of particulate matter mass from satellite AOT or in modules of global circulation and chemical transport models to compute aerosol radiative forcing effects, Hand and Malm, 2007). For convenience, model-derived MEE values are also included in Table 5.
3 Evaluation of the model performances and potential of its application

In this section, we evaluate the capability of the model results to reproduce ‘real’ aerosol conditions and explore the potential of the proposed model-based ALC inversion in producing quantitative geophysical information. In particular:

- In Section 3.1 we compare our simulations to real observations of independent backscatter and extinction coefficients made by different EARLINET Raman lidars (Bösenberg et al., 2001, Pappalardo et al., 2014).
- In Section 3.2, our model results are used to invert measurements acquired by some ALCs systems operating within ALICE-NET, which networks several ALCs systems (Nimbus CHM15k by Lufft) located across Italy and run by Italian research institutions and environmental agencies. Here we use data from some of these systems to derive the aerosol optical and physical properties (e.g. the aerosol optical thickness, AOT, and the aerosol volume and mass).

3.1 Comparison of the modelled aerosol optical properties to EARLINET measurements

As mentioned EARLINET Raman stations perform coordinated measurements two days per week following a schedule established in 2000 (Bösenberg et al., 2003). Overall, the EARLINET database includes the following categories: ‘climatology’, ‘CALIPSO’, ‘Saharan dust’, ‘volcanic eruptions’, ‘diurnal cycles’, ‘cirrus’, and ‘others’ (forest fires, photo smog, rural or urban, and stratosphere). To be comparable to our results, we used EARLINET \( \beta_a \) and \( \alpha_a \) coefficients at 355 nm and at 532 nm within the quality assured (QA) ‘climatology’ category (Pappalardo et al., 2014).

However, note that additional data filtering was necessary to screen out residual, likely unreliable values within this QA ‘climatology’ category. In particular, we only selected those EARLINET QA data further satisfying the following criteria:

- \( \beta_a \) and \( \alpha_a \) coefficients evaluated independently, i.e. only obtained using the Raman method (Ansmann et al., 1992);
- \( \beta_a \) and \( \alpha_a > 0 \);
- LR < 100;
- Relative errors on \( \beta_a \) and \( \alpha_a < 30\% \).

Then, we selected those sites in Europe expected to be mostly impacted by ‘continental’ aerosols and having the largest datasets (e.g., at least 100 points) at 355 and 532 nm. Overall, 5 sites satisfied these conditions (Table 6), and namely Madrid (Spain), Potenza and Lecce (Italy), Leipzig and Hamburg (Germany). Finally, being interested in continental conditions here, we filtered out those measurements dates affected by desert dust at the measuring sites, i.e. we removed from our ‘model-measurement comparison data set’ all the dates within the EARLINET ‘climatology’ category also belonging to the EARLINET ‘Saharan dust’ category.

Figure 4 depicts the results of the model-measurements comparison at the sites fulfilling our requirements in terms of LR vs \( \beta_a \) at \( \lambda=355 \text{ nm} \) (the corresponding results at \( \lambda=532 \text{ nm} \), including Madrid in place of Hamburg, are given in Appendix C, Figure C1). The colored area represents the model-simulated data range, while the color code indicates the absolute number of simulated values (i.e. counts) in each \( \beta_a \) - LR pair. The EARLINET-measured values are reported as black open circles. Note that, being the model simulations performed over an altitude range 0-5 km (see Section 2.1) only those simulations corresponding to the altitude range (\( \Delta z \)) covered by the measurements at each EARLINET station was taken into account here. Figure 4 shows the model results to well encompass the measured LR vs \( \beta_a \) data, with few measurements outside the modeled range (most of the exceptions are found for Potenza). Statistically, the
highest number density of simulated data well fits the observations, with the exception of Hamburg (Figure 4a), which however has the lowest number of measured data (it is not an EARLINET station any longer, see Table 6).

In Figure 5 the previous results at λ=355 nm are converted in terms of ‘mean’ LR per bin of βs for both model (blue) and observations (red, again, only βs bins containing at least 1% of the total modeled data were considered). This view shows that there is a general good agreement between the modeled and the measured LR values, and in their variation with βs. Some major deviations are found for Potenza and are further discussed in the following. The model-measurements accordance shown in Figure 5 was evaluated in quantitative terms by computing mean LR relative differences at both λ = 355 and 532 nm, i.e., we derived (LRmod - LRmean)/LRmean*100 values, where LRmod and LRmean are the lidar ratio values computed by model and derived by lidar measurements, respectively. These values are reported in Table 7 for each considered EARLINET station, together with the measurements-based mean LR in each observational site (computed weighting the number of observations per βs-spaced bins).

Results in Figure 5 and Table 7 also give some hints on the capability of the aerosol type assumed (and its admitted ranges of variability) to reproduce ‘real’ continental aerosol conditions in different sites across Europe. In fact, the four continental sites selected with our criteria are still expected to be partially impacted by different aerosol types.

- A good agreement between the model and the observations in terms of LR mean values is found for Hamburg (Figure 5a), with mean LR differences of the order of 5% (Table 7). Still, the measured LR values have a high variability and their distribution is positioned towards high values of βs (1×10\(^{-3}\) to 4×10\(^{-3}\) km\(^{-1}\) sr\(^{-1}\)). This could be due to the presence of different aerosol types as slightly polluted marine and polluted aerosol (Matthias and Bösenberg, 2002).

- A good accord for Leipzig (Fig. 5c) also indicates that this site is mostly dominated by ‘pure’ continental particles. In fact, the distribution of observed LR points in Fig. 4, which covers βs values ranging from 2×10\(^{-4}\) to 3×10\(^{-3}\) km\(^{-1}\) sr\(^{-1}\), is well centered to the modeled simulations highest density (counts > 40). Table 7 shows that at both wavelengths mean discrepancies with LR measurements keep well below 10%.

The highest differences in Fig. 5 are found in some southern Europe EARLINET sites:

- In Lecce (Fig. 5b), the best agreement between model and observations is found for the lowest values of βs (between 9×10\(^{-4}\) to 1×10\(^{-3}\) km\(^{-1}\) sr\(^{-1}\), see Table 7). Also, the increase from 10% to 18% in the discrepancies at 355 and 532 nm indicates some model problems in correctly reproducing the spectral variability of the optical properties, suggesting some mismatch between modeled and real aerosol sizes in this site (see discussion below).

- In Potenza (Fig. 5d), a significant difference between the mean LR curves emerges for βs values > 6×10\(^{-4}\) km\(^{-1}\) sr\(^{-1}\), with observed LR values lower than those simulated here.

These discrepancies could be due to the influence of marine aerosols at both stations (De Tomasi et al., 2006, Mona et al., 2006, Madonna et al., 2011), which is expected to produce lower LR values for high values of βs (e.g. BG01). In fact, Madrid shows better performances, with dLR/LR comparable to those in Leipzig.

To provide some insight into the reasons of the model-measurements differences at LC and PO sites, some specific model sensitivity tests have been performed and are reported in Appendix D. In particular, for Lecce, we found that better agreement between the observed and simulated LR vs βs behavior at 355 nm is obtained by reducing the variability range of N\(_{\text{tot}}\) (from 500 - 10000 cm\(^{-3}\) to 500 - 5000 cm\(^{-3}\) at ground). This indicates that LC is likely affected by cleaner continental aerosol type conditions. The sensitivity simulations done for understanding the mismatches with Potenza measurements show that an extension of the variability range of the coarse mode radius is needed to reproduce
the observed decrease of LR for increasing backscatter (Figure 5d). This suggests the presence of coarse particles larger than those assumed in such clean continental environment (Appendix D). This is compatible with the suspect of marine air contamination, although at this stage we are not able to exclude additional contamination of coarser particle of soil origin.

Overall, mean LR differences between our ‘average-continental’ model and data at selected continental sites in Europe keep lower than 20% (Table 7), this indicating it reasonably well reproduces the clean-to-moderately polluted continental aerosol conditions we intended to simulate.

3.2 Model results application to Nimbus CHM15-k ALC measurements

To test and validate the model-based inversion methodology, we used the derived functional relationships (Section 2.2) to invert and analyze the measurements of some ALICENET ALCs (Luft CHM15k systems). These instruments are biaxial ceilometers that emit laser pulses at 1064 nm (Nd:YAG-laser, class M1) with a typical pulse energy of 8 µJ and a pulse repetition rate of about 6500 Hz. The instruments have a specified range of 15 km and full overlap at around 1500 m (Heese et al., 2010). The manufacturer provides the overlap correction functions \( O(z) \) for each system. As shown recently by Wiegner and Geiß (2012) and Wiegner et al. (2014), a promising strategy to retrieve the aerosol backscatter coefficient from ALC measurement is adopting the forward solution of the Klett inversion algorithm (Klett 1985). This solution requires a known calibration constant of the system (i.e. absolute calibration, \( c_L \)) and an assumption on the LR. The advantage with respect to the backward solution is that calibration is not affected by the low SNR in the upper troposphere and it is needed occasionally. Furthermore, starting close to the surface, the data retrieval allows resolving aerosol layers in the boundary layer even if their optical depth is high. The forward solution of the Klett inversion algorithm is thus adopted here. For convenience, we report here the equations used within our procedure to obtain \( \beta_a \) from ALC measurements, which are also described in Wiegner and Geiß (2012, equations 1–3):

\[
\beta_a(z) = \frac{z(z)}{LR N(z)} - \beta_m(z) \tag{11}
\]

with

\[
Z(z) = LR z^2 P(z) \exp \left[ -2 \int_0^z (LR \beta_m - \alpha_m) dz' \right] \tag{12}
\]

and

\[
N(z) = c_L - 2 \int_0^z Z(z')dz'. \tag{13}
\]

Here, \( \beta_m \) and \( \alpha_m \) are the molecular backscatter and extinction coefficients calculated from climatological, monthly air density profiles and \( z^2 P(z) \) is the ALC range \( (z) \) corrected signal \( (P) \) (also referred to as RCS), that is the raw data obtained by the considered ALCs. As anticipated, knowledge of the calibration constant \( c_L \) is needed to solve eq. 13 (and thus 11, forward solution). In our analysis of ALC daily records, the constant \( c_L \) has been obtained by the “backward approach” (Rayleigh calibration) applied to night-time, cloud-free ALC signal averaged over 1 or 2 hours at 75 m height resolution. This allows for using the best \( c_L \) retrieval (that is the night-time, lowest noise one), in the forward solution of the lidar equation, which guarantees operating over the best signal to noise range of the ALC signal.
3.2.1 Model-based retrieval of aerosol optical properties

Operatively, inversion of the aerosol properties, $a_0(z)$ and $\beta_0(z)$, is performed using an iterative technique, since we need to correct the backscatter signal at each altitude $z$ for extinction losses. The iterative procedure is stopped when convergence in the integrated aerosol backscatter ($\text{IAB} = \Sigma_0 \cdot a_0(z)$) is reached (e.g. BG01). At each step, aerosol extinction is derived using the functional relationship $a_0 = a_0(\beta_0)$ of Table 3.

An example of the outcome of this retrieval methodology is depicted in Figure 6. It shows the time-height (24h, 0 - 6 km) contour plot of $a_0$ retrieved at 1064 nm during a whole day of measurements (June 26, 2016) performed by the ALICENET system of Aosta San Christophe (ASC, 45.8°N, 7.4°E 570 m a.s.l., Northern Italy, Figure 7a). Time and altitude resolutions are 1 min and 15 m, respectively. Note that ALC data are cloud-screened using the cloud mask of the Lufft firmware.

The aerosol optical thickness (AOT) is obtained vertically integrating the ALC-derived $a_0(z)$ from the surface up to a fixed height $z_{\text{AOT}}$, above which the aerosol contribution is assumed to be negligible. In Figure 6, the ALC-derived AOT values at 1064 nm (pink curve, with a temporal resolution of 5 min) is superimposed to the extinction contour. Reference AOT values from a co-located sun-sky radiometer (a Prede POM-02 system) are shown by orange circles. These were extrapolated at 1064 nm from the instrument 1020 nm-channel using the Angström exponent derived fitting AOT values at all the radiometer wavelengths. This example illustrates the very good performances of our model-assisted inversion scheme, and the capability of this approach to extend to nighttime the (daylight-only) radiometer observations.

To evaluate the performances of our model-assisted retrieval of $a_0(z)$ over a more statistically significant dataset, the same approach illustrated in Figure 6 was applied to a longer record in the ASC site, plus Nimbus CHM-15k ALC datasets from two additional ALICENET sites: San Pietro Capofiume (SPC, 44°39N, 11°37E, 10 m a.s.l.) and Rome Tor Vergata (RTV, 41.88°N, 12.68°E, 100 m a.s.l.). The location of the instruments is shown in Figure 7a (red circles), while some information on system types and site characteristics is given in Table 8. The data analyzed here were collected during the following periods: April 2015 – June 2017, June 2012 – June 2013 and February 2014 – September 2015, for ASC, SPC and RTV, respectively.

In those sites, reference AOTs were collected by three co-located sun-sky radiometer, and namely using two SKYNET Prede sun-sky radiometers at ASC and SPC (POM-02L and POM-02, respectively, www.euroskyrad.net) and an AERONET Cimel CE 318-2 instrument operational at RTV (https://aeronet.gsfc.nasa.gov, Rome Tor Vergata station, data level 2.0). Only AOT values between 0.01 and 0.2 at 1064 nm were considered. This range allows for excluding the data points with 1064 nm-AOT lower than the sunphotometer expected accuracy ($\text{dAOT} = 0.01$) and those where we found aerosol extinction to cause significant deterioration of our ALC signal. Overall a total of 1237, 268, 850 AOT pairs were analyzed at ASC, SPC and RTV, respectively.

Also note that, although CHM-15k data are already corrected for the $O(z)$ function provided by the manufacturer, the variation of the ALC internal temperature was shown to lead to $O(z)$ differences up to 45% in the first 300 m above ground (Hervo et al., 2016). For this reason, in our analyses the lowest valid altitude of the CHM-15k for both the SPC and RTV systems was fixed to be about 400 m. A linear fit of the first two valid ALC points is then used to extrapolate $a_0(z)$ down to the ground ($z_0$). Conversely, due to the optimal characterization down to the ground of $O(z)$ provided by Lufft for the CHM-15k system installed at ASC, values at $z_0$ at this site are not those extrapolated but actually those.
measured. The maximum altitude of aerosol extinction vertical integration to derive the AOT, $z_{\text{AOT}}$, was selected as the first height above 4000 m where the range corrected signal (RCS) has a SNR < 1.

Results of the long-term AOT comparison are summarized in Figure 7 and Table 9. For each site under investigation, Figure 7 shows the histograms of the AOT differences between the hourly-mean coincident AOTs as derived by the ALCs and measured by the sun-photometers (red curve, corresponding AOT vs AOT scatter plots at the three considered sites are given in Appendix E). To evaluate the advantage of our approach with respect to more standard lidar inversions, we also computed AOT differences using two fixed-LR values. In particular, we used LR = 52 sr (i.e. the value suggested by the E-Profile network, black lines) and LR = 38 sr (i.e. the weighted mean LR value derived from our model, see Section 3, blue lines). Figure 7 shows that the best agreement is found at ASC. The distribution of AOT difference has a maximum around 0 for each of the three inversions schemes, with very low dispersion. The full width at half maximum, FWHM, is in fact around 0.015, and approximately 55% of the data are included in the interval -0.01 – 0.01, which is even within the expected error of photometric measurement. For SPC and RTV, the red and blue histograms are peaked around 0, whereas the black ones are shifted, with maxima around 0.01-0.02 and 0.02-0.03 for SPC and RTV, respectively. These two sites have higher dispersion (FWHM = 0.03), and approximately 30% of the data are included in the interval -0.01 – 0.01 for the red and blue histograms at both sites, which is probably due to the different aerosol load affecting the different ALICENET stations. As pointed out by the low value of the average AOT computed at ASC for the analyzed dataset ($<\text{AOT}> = 0.027$), low pollution levels generally characterize this site, with some exceptions due to wind-driven aerosol transport from the nearby Po valley (Diémoz et al., 2018a, 2018b this issue).

On the contrary, RTV ($<\text{AOT}> = 0.044$) and, especially, SPC in the Po Valley ($<\text{AOT}> = 0.076$) are characterized by higher aerosol content and pollution levels, which explain the larger histogram dispersions. Note that the high frequency of fog events in winter markedly reduces the number of analyzed AOT pairs at SPC site, while some desert dust affected days at both SPC (e.g., Bucci et al., 2018) and RTV (e.g., Barnaba et al., 2017) were removed from our datasets (no desert-dust affected dates in ASC).

Table 9 summarizes the long-term performances of the model-based procedure in deriving quantitative AOT from the ALC systems at the three investigated sites. It includes values of the average differences between the ALC-derived and sun-photometers-measured AOT (both bias, $<\Delta\text{AOT}>$, and absolute difference $|<\Delta\text{AOT}>|$, with associated standard deviations) obtained using both the proposed model-based approach and the fixed-LR inversions. For SPC and RTV sites, these numbers show that the best ALC–photometer accordance is reached when employing either the model-based or the fixed LR=38 sr inversion scheme. In fact, these two approaches have similar performances in terms of mean $\Delta\text{AOT}$ values ($<|\Delta\text{AOT}|> = 0.011, 0.013$ and 0.013, 0.014 for SPC and RTV, respectively), mean percent error ($<|\Delta\text{AOT}|>/<\text{AOT}> = 0.16, 0.19 and 0.31, 0.33$) and a very low mean relative bias ($<\Delta\text{AOT}>/<\text{AOT}> = -0.043, 0.005$ and 0.088, 0.11). On the other hand, the fixed LR=52 sr retrieval produces an overestimation of AOT in both SPC and RTV ($<|\Delta\text{AOT}|>/<\text{AOT}> = 0.33$ and 0.44) with larger discrepancies between retrieved and observed AOTs ($<|\Delta\text{AOT}|> = 0.021$ and 0.026, $<|\Delta\text{AOT}|>/<\text{AOT}> = 0.38$ and 0.49). For the ASC site, due to the low aerosol content, the differences among the inversion schemes are almost negligible.

Overall, for the three sites, the statistics over the long-term datasets employed showed good results of the model-based approach with similar behavior of the retrievals with a fixed LR of 38 sr, while a fixed LR value of 52 sr produces an overestimation of the AOT at SPC and RTV. As different sites have different (and not known a-priori) characteristic LR values, these results highlight the potential of the model-based approach to derive quite accurate $\beta_s$ and $\alpha_s$ coefficients without the need to choose and fix an arbitrary LR value.
3.2.2 Model-based retrieval of aerosol volume (and mass)

In this section we provide examples of the applicability of the proposed approach to derive air-quality relevant parameters. In particular, we use the ALC, $B_\alpha$-retrieved data and the 7th-order polynomial fit linking $B_\alpha$ (at $\lambda = 1064$ nm) to $V_a$ (see also Table 3 and Figure 2c) to derive the aerosol volume (and mass).

The ALC-estimates were firstly compared to aerosol volume derived in situ at the ASC site by two different optical particle counters (OPCs) on 29th December 2016 and 5th September 2017. For the case of the 29th December 2016, a TSI Optical Particle Sizer (OPS) 3330 was employed. This instrument has 16 channels that can be programmed to provide the number concentration at different (and logarithmically spaced) diameter size ranges within the interval 0.3 - 10 $\mu$m. Further details can be found in the TSI manual (2011). For the case of the 5th September 2017, the Fidas®200s OPC was used. This spectrometer is able to retrieve high-resolution particle spectra (size measurements between 0.15 and 27 $\mu$m, with 32 channels/decade, Pletscher et al., 2016). For both dates, Figure 8 shows the time (x-axis, 24h) vs. height (left y-axis) contour plots of the ALC-based retrieval of the aerosol volume concentration (cm$^{-3}$). The OPC-derived aerosol volume concentration measured at ground-level is reported as a function of time (x-axis) on the right y-axis (grey curve). The corresponding ALC-derived volume concentration (integrating the ALC data between 0 and 75 m) is shown by a pink curve (same right y-axis). Daily mean volume concentration values derived by OPCs and by ALC are also plotted (grey cross and pink triangle, respectively). The horizontal bar in the upper part of the figure indicates the ranges of RH measured in-situ during the analyzed cases.

The OPC-to-ALC comparison is certainly affected by intrinsic factors, as differences on the atmospheric layer sampled (at ground and integrated between 0 and 75 m, for OPC and ALC, respectively) and on the probing methods (in-situ and remote sensing, dried air sampled by OPC and ambient conditions sampled by the ALC). Furthermore, as mentioned in Section 4.2.1, a major critical issue of ALC retrievals at low levels is the correction for the overlap function, which needs to be experimentally characterized and verified for each instrument.

These issues are visible in the given example of Figure 8. In fact, in the upper panel, the agreement between the ALC-derived and the TSI-OPC aerosol $V_a$ values is good between 0 and 7 UTC. In the following hours both instruments register an increase of the aerosol volume, although with some discrepancies in absolute values. Starting from 18 UTC, the ALC derives an aerosol volume concentration higher than the OPC one by a factor of 2-3.5. This disagreement could be related to both the presence/arrival of fine particles (<0.3 $\mu$m) not measured by the optical counter (see for example Diémoz et al., 2018a), or to aerosol hygroscopic effects (increase of volume associated to hygroscopic growth seen by ALC but not by the OPC which dries the air samples). This latter effect is confirmed by the large RH values (RH > 90%) measured after 18 UTC. The lower panel shows a good agreement between the ALC-derived and the Fidas OPC $V_a$ values, in particular until 4 UTC and after 16 UTC. Some differences emerge around 7 UTC and between 11 and 15 UTC, where the ALC volume is lower by a factor of 2 compared to the in situ Fidas $V_a$ values. The smaller minimum detectable size of the Fidas OPC instrument with respect to the OPS is likely the reason for the better accord between ALC and OPC $V_a$ values in this test date. In this case, the effect of RH seems to be less important, and indeed RH values keep lower than 90%.

In general, high RH values (RH >= 90%) are known to markedly affect the aerosol mass estimation from remote sensing techniques and its relationship with ‘reference’ PM2.5 or PM10 measurements methods, usually performed in dried conditions (e.g. Barnaba et al., 2010; Adam et al., 2012, Li et al., 2016, Li et al., 2017). This theme is also discussed in Diémoz et al. 2018a for the ALC measurement site of Figure 8. Nevertheless, even with the mentioned
limitations, results in Fig. 8 well show the potential of the developed method in providing sound values of aerosol volume, and hence, mass, in average-RH regimes, giving support to more standard PM10 air quality monitoring.

To give a further example in this direction, the model-assisted retrievals of aerosol mass over a longer time period were used to derive daily-mean aerosol mass concentrations (PM10), a measurement typical of air quality stations. To this purpose, for the two-months period June-July 2012, we derived daily mean values of aerosol volume at the SPC site using the functional relationships \( V_a = V_a(p_a) \), and then converted these into mass (PM10) using typical values of aerosol densities \( \rho_a \). Results are shown in Figure 9. It compares the daily average PM10 concentration measured in situ at SPC by the Italian Regional Environmental Protection Agency (ARPA, red solid curve) and the model-assisted, ALC-derived daily mass concentration obtained assuming both a fixed particle density \( \rho_a = 2 \ g/cm^3 \) (blue dotted curve), and a range of it between 1.5-2.5 g/cm³ (shaded area), this range covering approximately the typical \( \rho_a \) values at the SPC site. Yellow shaded areas indicate the presence of dust events (e.g. Bucci et al., 2018) that are excluded from the results reported in the next paragraph.

More in detail, the daily-mean, ALC-derived mass concentrations were estimated in two steps: 1) estimation of hourly mass values for the selected height; 2) computation of the daily values through the median of the hourly values. To guarantee a good daily representativeness, the second step is applied only to those days in which at least 50% of the hourly values is available in all the following temporal ranges: 00 - 05 UTC, 06 - 11 UTC, 12 - 17 UTC, 18 - 23 UTC.

Note that, due to the uncertainties associated to the modelled aerosol properties (e.g., particle density), the uncertainty in the ALC retrievals is expected to be only slightly affected by this height difference, particularly in daylight, due to the strong convection within the mixing layer. Possible exceptions could be in nocturnal conditions when vertical gradients in the lowermost hundreds of meters can occur. However, our statistical (3-year) ALC records show the mixing layer height at SPC to descend below 250 m only 4-5 hours per day in July (usually between 22 and 3 UTC, i.e., when emissions are at a minimum). Overall, Figure 9 confirms a good agreement between the ALC-derived and the ARPA reference PM10 values, with a correlation coefficient (R) of 0.64. In fact, mean, absolute mean and relative differences, between the two series are: \( \langle d\text{PM10} \rangle = 2.3 \pm 6.0 \ g/cm^3 \), \( \langle d\text{PM10}/\text{PM10} \rangle = 0.14 \pm 0.27 \). This agreement attests that SPC site can indeed be considered an ‘average’ continental site and suggests the potential of this approach to derive information on aerosol volume and mass. Still, due to the specificity of each site and to the limited period considered here, these results cannot be taken as representative of all continental sites at all times. Further studies at different places and over longer time periods would be necessary to better assess the uncertainty of the proposed retrieval, including uncertainties due to the variability of “continental” conditions (in terms of particle size distribution, compositions, hygroscopic effects, etc...), but also of the instrument-dependent performances (e.g. overlap corrections, etc...).

4 Summary and Discussion

Thanks to their low construction/operation costs and to their capability at providing continuous, unattended measurements, the use of automated-lidar-ceilometers (ALCs) for aerosol characterization has increased in the recent years. Several numerical approaches were recently proposed to estimate the aerosol vertical profile either using ceilometer measurement only, or coupling these with ancillary measurements (e.g., Stachlewska et al., 2010; Flentje et al., 2010; Wiegner et al., 2012; Wiegner et al., 2014; Cazorla et al., 2017, Román et al., 2018).
This work proposes a methodology to retrieve key aerosol properties (as extinction coefficient, surface area and volume, thus mass) from lidar/ALC measurements using in support the results from a specifically developed aerosol numerical model to drive the retrievals. In particular, the numerical model uses a "Monte-Carlo" approach to simulate a large set (20000) of aerosol microphysical properties intended to reproduce the variability of 'average' (clean-to-moderately polluted) continental conditions, i.e., those expected to dominate over Europe. Based on the assumption of particle sphericity, relevant computations of aerosol physical (surface area and volume, $S_a$ and $V_a$) and optical (backscattering and extinction coefficients, $\beta_a$ and $\alpha_a$ through Mie scattering theory) properties were performed at three commonly used lidar wavelengths (i.e., at the Nd:YAG laser harmonics 355, 532, 1064 nm). Fitting procedures of this large set (20,000) of $\beta_a$ vs. $\alpha_a$, $S_a$ and $V_a$ data-pairs were then used to derive mean functional relationships linking $\beta_a$ to $\alpha_a$, $S_a$ and $V_a$, respectively. The model’s statistical uncertainties (i.e., those related to the variability of the microphysical parameters used in input to the computations of the bulk physical/optical properties) associated to these so-derived mean relationships were found to be within 30% and 40% for $\beta_a$ vs $\alpha_a$ and $\beta_a$ vs $V_a$, respectively, while $\beta_a$ vs $S_a$ exhibits a larger dispersion (relative standard uncertainty of 40%-70%, depending on $\beta_a$). It is worth mentioning that these are higher than those associated to the retrievals of aerosol bulk parameters using the complete set of Raman lidar observations (three aerosol backscattering and two extinction coefficients, i.e., the so called 3+2 approach) assuming, as in our case, no random uncertainty in the lidar input data. For example, Veseloski et al. (2012) found a maximum uncertainty of 15% for particle volume and surface area estimation, in the case of 0% random uncertainty in the lidar input data. Note however, that such multi-wavelength lidar systems need to be operated by highly trained operators, and are rarely run all day round.

The model results also allowed exploring the expected dependence of the (continental aerosol) lidar ratio (LR) on $\beta_a$ at 355, 532 and 1064 nm, and in turn, the mean, 'weighted' -LR value at each wavelength (found to be 50.1 ± 17.9 sr, 49.6 ± 16.0 sr and 37.7 ± 12.6 sr, at 355, 532 and 1064 nm respectively). Availability in literature of LR values at 1064 nm are scarce and its monotonic increase with $\beta_a$ found in this work (Figure 3) suggests that the use of a fixed LR value for the inversion of ALC signals should be done with caution and carefully evaluated case by case. A similar, non-monotonic behavior characterizes the shapes of LR vs $\beta_a$ curve at 355 and 532 nm.

We tested the reliability of our model results in two ways: 1) the model numerical computations were compared to 'real' lidar measurements (specifically selected within the EARLINET database), and 2) the model-assisted retrievals of aerosol optical (AOT) and physical ($V_a$, PM10) properties by real, operational ALC systems were compared to corresponding 'reference' measurements performed by co-located, independent instrumentation.

In particular, in task 1) our simulations were compared to backscatter and extinction coefficients at 532 and 355 nm independently retrieved by advanced Raman lidar systems operating at different EARLINET sites in Europe (namely Hamburg and Leipzig in Germany, Madrid in Spain, Lecce and Potenza in Italy). The model simulations were found to statistically well match the observations (Figures 4, 5 and C1). Mean discrepancies between model and measurement-based LR were found to be lower than 20%, suggesting a good capability of the assumed aerosol model (and admitted range of variability) to represent 'real', 'average continental' aerosol conditions in different sites across Europe. Some differences emerged for the southern Italy EARLINET sites, possibly affected by the influence of marine aerosols, leading to lower LR values for high values of $\beta_a$.

For task 2) we applied the proposed model-based inversion to different ALC systems (Lufft CHM-15k), part of the Italian ALICENET network. We firstly tested the ability of the proposed approach to derive aerosol extinction by comparing hourly-mean, vertically-integrated $\alpha_a$ (i.e., hourly mean AOT) derived by three ALC systems to corresponding AOT measurements from co-located sun-photometers (ALICENET sites of Aosta San Cristophe (ASC),
San Pietro Capofiume (SPC) and Rome Tor Vergata (RTV), Figure 7. ALC-sun photometer agreement was found to be within 30%. Tests on the use of fixed LR were also performed to investigate the advantage of the proposed approach with respect to more standard ones. To this purpose, we used the (1064 nm) fixed-LR value suggested by the E-Profile EUMETSAT Program and the ‘weighted mean’ derived from our model (52 sr and 38 sr, respectively). While for the ASC site negligible differences were found among the three retrieval schemes, for both SPC and RTV sites the best ALC – sun photometer accordance in AOT is reached when employing the model-based or the fixed LR=38 sr inversion schemes, with a mean error around 16-19 % and 31-33 % for SPC and RTV, respectively. Applying the fixed LR value of 52 sr produces an overestimation of the AOTs, with a mean relative bias equal to 33 % and 44 % at SPC and RTV, respectively. This suggests that, at 1064 nm, the LR value for continental aerosol is lower than the one assumed by the E-Profile procedure and, more in general, this highlights the advantage of a procedure not requiring an a-priori, and to some extent arbitrary, choice of the LR value.

As a second test in task 2, values of aerosol volume (and mass) derived using the model-assisted ALC retrieval were compared to in situ aerosol measurements performed by OPcs and PM10 analyzers. A continuous, two-months comparison (June – July 2012) between daily average aerosol mass concentration as measured in situ and derived by ALC (in the lowest altitudes) at SPC, showed a mean relative difference of around 15% (Figure 9).

Overall, the good results obtained in our validation efforts are encouraging but necessarily related to the specific conditions at the measuring sites considered and to the characteristics of the instruments employed. They are therefore not necessarily representative of results obtainable in all European continental sites, and at all times. Further tests using wider datasets covering a variety of sites and ALC instrumentation would be desirable to better understand potential limits of the applicability of the proposed method over the larger scale. An obvious intrinsic limitation is that the method is dependent on the considered aerosol type which in this study was tuned to reproduce average continental aerosol conditions. Errors associated to the application of the derived functional relationship might be larger if more ‘specific’ aerosol conditions (e.g. contamination by sea salt or desert dust particles) affect a given site. In the future, the information coming from ALC systems with an additional depolarization channel (as tested in the DIAPASON Project, Gobbi et al., 2018) could be used to force the retrieval to different model schemes (e.g. switching from ‘no dust’ to ‘dust’ schemes conditions) in the same vertical profile. This will enhance the capabilities of ALCs to operatively estimate and characterize the aerosol optical properties (e.g. Gasteiger and Freudenthaler, 2014).

Additionally, although our validation exercises returned results well within the uncertainties related to the model statistical variability alone (i.e., the relative errors associated to the mean functional relationships), the expected total uncertainty to be associated to the method should include terms that have not been specifically addressed in this work, as for example the instrumental error itself.

On the other hand, the proposed approach has the main advantage of allowing the operational (i.e. 24/7) retrieval of fairly reliable, remote sensing profiles of aerosol optical ($\beta_0$, $\alpha_0$) and physical ($S_a$, $V_a$) properties (with associated uncertainties and limitations) by means of relatively simple and robust instruments. This could temporally and spatially complement the information coming from more advanced lidar networks (for example, the Raman channel of multi-wavelength system cannot be used in daylight conditions) and, more in general, could represent a valid option to deliver, in quasi real time, the 3D aerosol fields useful for operational air quality (e.g. integration of the in situ surface measurements) and for meteorological and climate monitoring (e.g. aerosol-cloud interaction and aerosol transport and dispersion processes).
Acknowledgements

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Data availability

AERONET Rome-Tor Vergata sun photometer AOT data were downloaded from the AERONET web page (AERONET, 2018). SKYNET sun photometer AOT data were downloaded from the SKYNET webpage (SKYNET, 2018). EARLINET backscattering and extinction coefficients were downloaded from the EARLINET webpage (EARLINET, 2018). ALICENET ALC raw data are available upon request at alicenet@isac.cnr.it.


time processing of a ceilometer network assisted with sun-photometer data: monitoring a dust outbreak over the Iberian Peninsula, Atmos. Chem. Phys., 17, 11861-11876, https://doi.org/10.5194/acp-17-11861-2017, 2017


Hanel, G.: The properties of atmospheric aerosol particles as function of the relative humidity at thermodynamic equilibrium with the surrounding moist air, Adv. Geophys., 19, 73–188, 1976


TSI, Model 3330 optical particle sizer spectrometer operation and service manual, P/N 6004403, Revision E, April 2011.


Table 1. Aerosol parameter values as reported in literature for continental-type aerosols.

<table>
<thead>
<tr>
<th>Reference</th>
<th>( r_1 ) (µm)</th>
<th>( r_2 ) (µm)</th>
<th>( r_3 ) (µm)</th>
<th>( N_1/N_{tot} )</th>
<th>( N_2/N_{tot} )</th>
<th>( N_3/N_{tot} )</th>
<th>( m_{r,1} )</th>
<th>( m_{r,2} )</th>
<th>( m_{r,3} )</th>
<th>( m_{im,1} )</th>
<th>( m_{im,2} )</th>
<th>( m_{im,3} )</th>
<th>( N_{tot} )</th>
<th>Aerosol type</th>
</tr>
</thead>
<tbody>
<tr>
<td>Whitby (1978)</td>
<td>0.008</td>
<td>0.034</td>
<td>0.46</td>
<td>0.56</td>
<td>0.44</td>
<td>( 4 \times 10^4 )</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>1800</td>
</tr>
<tr>
<td></td>
<td>1.6</td>
<td>2.1</td>
<td>2.2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Clean continental</td>
</tr>
<tr>
<td>D'Almeida et al. (1991)</td>
<td>0.012</td>
<td>0.029</td>
<td>0.471</td>
<td>0.06</td>
<td>0.94</td>
<td>( 2 \times 10^5 )</td>
<td>1.75</td>
<td>1.53</td>
<td>1.53</td>
<td>( 0.44 )</td>
<td>0.012</td>
<td>0.008</td>
<td>20000</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.0</td>
<td>2.24</td>
<td>2.51</td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Average continental</td>
</tr>
<tr>
<td>Hess et al. (1998)</td>
<td>0.012</td>
<td>0.021</td>
<td>0.471</td>
<td>0.56</td>
<td>0.44</td>
<td>( 0.3 \times 10^4 )</td>
<td>1.75</td>
<td>1.53</td>
<td>1.53</td>
<td>( 0.44 )</td>
<td>0.012</td>
<td>0.008</td>
<td>15300</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.0</td>
<td>2.24</td>
<td>2.51</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Average continental</td>
</tr>
<tr>
<td>Barnaba and Gobbi (2004a)</td>
<td>0.007–0.021</td>
<td>0.040–0.59</td>
<td>6.1–54.2</td>
<td>65.8</td>
<td>12.5–20.0</td>
<td>1.53–1.57</td>
<td>( 0.7 \times 10^4 )</td>
<td>6 \times 10^3</td>
<td>( 8 \times 10^3 )</td>
<td></td>
<td></td>
<td></td>
<td>( 10^7 \cdot 10^9 )</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>0.012</td>
<td>0.077</td>
<td>0.5</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Clean and polluted continental</td>
</tr>
<tr>
<td>Omar et al. (2009)</td>
<td>1.7–2.0</td>
<td>2.03–2.11</td>
<td>2.24–2.24</td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td></td>
<td>-</td>
<td>0.093–0.10</td>
<td>0.68–0.76</td>
<td>0.999–1</td>
<td>(0.02–3)</td>
<td>-</td>
<td>( 1.38 \cdot 10^4 )</td>
<td>( 0.1–6.3 )</td>
<td>(3.4–6.3)</td>
<td>( 0.01–0.02 )</td>
<td>-</td>
<td>-</td>
<td>-</td>
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<tr>
<td></td>
<td>-</td>
<td>1.53–1.61</td>
<td>1.9–2.1</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td>Clean and polluted continental</td>
</tr>
<tr>
<td>Levy et al. (2007)</td>
<td>0.018</td>
<td>0.005</td>
<td>0.5</td>
<td>1</td>
<td>( 1 \times 10^7 )</td>
<td>( 1 \times 10^{17} )</td>
<td>1.75</td>
<td>1.53</td>
<td>1.53</td>
<td>( 2 \times 10^{10} )</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.0</td>
<td>2.97</td>
<td>2.97</td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Barnaba et al. (2007)</td>
<td>-</td>
<td>0.05–0.1</td>
<td>0.4–0.5</td>
<td>0.98–0.99</td>
<td>0.01–0.02</td>
<td>-</td>
<td>( 1.35 \cdot 15.5 )</td>
<td>( 1.53 \cdot 16 )</td>
<td>( 1 \cdot 10^3 )</td>
<td>( 2.5–0.20 )</td>
<td>(1.0–80)</td>
<td>( 1 \cdot 10^3 )</td>
<td>Continental or coastal</td>
<td></td>
</tr>
<tr>
<td></td>
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<td>1.35–1.70</td>
<td>1.5–2.0</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Amiridis et al. (2015)</td>
<td>-</td>
<td>0.03–0.96</td>
<td>0.47–0.69</td>
<td>1</td>
<td>( 4–8 )</td>
<td>-</td>
<td>( 1.42 \cdot 14.5 )</td>
<td>( 1.45 \cdot 1.53 )</td>
<td>-</td>
<td>( 2.3–6 )</td>
<td>( 2.3–6 )</td>
<td>( 10 )</td>
<td>( 10 )</td>
<td>Clean and polluted continental</td>
</tr>
<tr>
<td></td>
<td>1.6–2.2</td>
<td>1.9–2.5</td>
<td>( 10^7 )</td>
<td>( 2.3 \cdot 10^7 )</td>
<td>( 2.3–6 )</td>
<td>-</td>
<td>( 2.3 \cdot 10^7 )</td>
<td>( 2.3–6 )</td>
<td>( 10^7 )</td>
<td>( 10^7 )</td>
<td>( 10 )</td>
<td>( 10 )</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

1 The refractive index is at \( \lambda = 532 \) nm.

2 The refractive index is at \( \lambda = 550 \) nm.
Table 2. Variability ranges used in this study. Values refer to ground and dry conditions (see text for details).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Mode I</th>
<th>Mode II</th>
<th>Mode III</th>
</tr>
</thead>
<tbody>
<tr>
<td>$r_i$ ($\mu$m)</td>
<td>0.005 - 0.03</td>
<td>0.03 - 0.1</td>
<td>0.3 - 0.5</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>1.35 – 1.7</td>
<td>1.35 – 1.7</td>
<td>1.5 – 2.4</td>
</tr>
<tr>
<td>$N/N_{tot}$ (%)</td>
<td>10 - 60</td>
<td>40 - 90</td>
<td>0.01 – 0.5</td>
</tr>
<tr>
<td>$m_i$ (355 nm)</td>
<td>1.40 – 1.80</td>
<td>1.40 – 1.70</td>
<td>1.50 – 1.60</td>
</tr>
<tr>
<td>(532 nm)</td>
<td>1.40 – 1.80</td>
<td>1.40 – 1.70</td>
<td>1.50 – 1.60</td>
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<tr>
<td>(1064 nm)</td>
<td>1.42 – 1.82</td>
<td>1.37 – 1.66</td>
<td>1.50 – 1.60</td>
</tr>
<tr>
<td>$m_{i, m}$ (355 nm)</td>
<td>$1 \times 10^{-2}$</td>
<td>$1 \times 10^{-4}$</td>
<td>$1 \times 10^{-4}$</td>
</tr>
<tr>
<td>(532 nm)</td>
<td>$1 \times 10^{-3}$</td>
<td>$1 \times 10^{-4}$</td>
<td>$1 \times 10^{-4}$</td>
</tr>
<tr>
<td>(1064 nm)</td>
<td>$1 \times 10^{-3}$</td>
<td>$1 \times 10^{-4}$</td>
<td>$1 \times 10^{-4}$</td>
</tr>
<tr>
<td>$N_{i, m}$ (cm$^{-3}$)</td>
<td>500 – 10000</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 3. Parameters of the Seventh-Order Polynomial Fits ($y = a_0 + a_1x + a_2x^2 + a_3x^3 + a_4x^4 + a_5x^5 + a_6x^6 + a_7x^7$) for $\lambda = 1064$ nm, with $x = \log(\beta_a)$ (in km$^{-1}$ sr$^{-1}$) and $y = \log(\alpha_a, S_a, or V_a)$ in (km$^{-1}$, cm$^3$/cm$^3$ and cm$^3$/cm$^3$, respectively).

<table>
<thead>
<tr>
<th>Functional relationship at 1064 nm</th>
<th>Extinction</th>
<th>Surface area</th>
<th>Volume</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a_0$</td>
<td>3.79737507651898</td>
<td>12.019452592845141</td>
<td>-5.314834128998254</td>
</tr>
<tr>
<td>$a_1$</td>
<td>3.294032541389781</td>
<td>30.825966279368547</td>
<td>2.500484347793244</td>
</tr>
<tr>
<td>$a_2$</td>
<td>0.962603336867675</td>
<td>24.518531616019207</td>
<td>-1.196109537503000</td>
</tr>
<tr>
<td>$a_3$</td>
<td>0.241796629870675</td>
<td>10.625241994796593</td>
<td>-1.583236058579546</td>
</tr>
<tr>
<td>$a_4$</td>
<td>0.064609145804688</td>
<td>2.634051072085453</td>
<td>-0.681801883947768</td>
</tr>
<tr>
<td>$a_5$</td>
<td>0.017721752150233</td>
<td>0.373150843707711</td>
<td>-0.145232662646142</td>
</tr>
<tr>
<td>$a_6$</td>
<td>0.002722551625862</td>
<td>0.027971628176431</td>
<td>-0.015471229968392</td>
</tr>
<tr>
<td>$a_7$</td>
<td>0.000157245409783</td>
<td>0.000854381337164</td>
<td>-0.000658925756875</td>
</tr>
</tbody>
</table>
Table 4. Mean weighted LR at 355, 532 and 532 nm derived in this work and comparison to the corresponding aerosol subtypes (clean continental, CC, and polluted continental, PC) from relevant literature.

<table>
<thead>
<tr>
<th>LR (sr)</th>
<th>λ=355 nm</th>
<th>λ=532 nm</th>
<th>λ=1064 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Omar et al., (2009) (Calipso aerosol model)</td>
<td>-</td>
<td>70 ± 25 (PC)</td>
<td>30 (PC)</td>
</tr>
<tr>
<td></td>
<td>-</td>
<td>35 ± 16 (CC)</td>
<td>30 (CC)</td>
</tr>
<tr>
<td>Amiridis et al. (2015) (LIVAS database)</td>
<td>59.5 (PC)</td>
<td>64 (PC)</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>56.5 (CC)</td>
<td>54 (CC)</td>
<td>-</td>
</tr>
<tr>
<td>Papagiannopoulos et al. (2016) (EARLINET measurements)</td>
<td>-</td>
<td>62 ± 10 (PC)</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>-</td>
<td>47 ± 4 (CC)</td>
<td>-</td>
</tr>
<tr>
<td>Düsing et al. (2018) (in-situ and lidar measurements)</td>
<td>55</td>
<td>55</td>
<td>30; 15°*</td>
</tr>
<tr>
<td>This work</td>
<td>50.1 ± 17.9</td>
<td>49.6 ± 16.0</td>
<td>37.7 ± 12.6</td>
</tr>
</tbody>
</table>

* derived using the extinction-related and backscatter-related Ångström exponents given by Amiridis et al. (2013)

** see the explanation in the text for the two different values
Table 5. Extinction-to-volume conversion factors, $c_v = V_a/\alpha_a$ (and corresponding ‘mass-to-extinctions efficiency’ values, MEE = $\alpha_a/(V_a \cdot \rho_a)$, given assuming $\rho_a = 2 \text{ g/cm}^3$) of continental particles as derived from our model at different wavelengths

<table>
<thead>
<tr>
<th>Reference</th>
<th>355</th>
<th>532</th>
<th>1064</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength [nm]</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>355</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>532</td>
<td>-</td>
<td>0.35 (1.43)</td>
<td>-</td>
<td>OPAC, clean continental model</td>
</tr>
<tr>
<td>1064</td>
<td>-</td>
<td>0.28 (1.79)</td>
<td>-</td>
<td>Opac, polluted continental model</td>
</tr>
<tr>
<td>Hess et al. (1998)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Barnaba and Gobbi (2004b)</td>
<td>-</td>
<td>0.18 (2.78)</td>
<td>-</td>
<td>Continental model</td>
</tr>
<tr>
<td>Ansmann et al. (2011b)</td>
<td>-</td>
<td>0.18 (2.78)</td>
<td>-</td>
<td>Germany, fine aerosol fraction</td>
</tr>
<tr>
<td>Lewandosky et al. (2010)</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>Mexico city basin</td>
</tr>
<tr>
<td>Sicard et al. (2012)</td>
<td>-</td>
<td>0.26 (1.92)</td>
<td>-</td>
<td>AERONET, Spain</td>
</tr>
<tr>
<td>Mamouri and Ansmann (2017)</td>
<td>0.17 (2.94)</td>
<td>0.30 (1.67)</td>
<td>0.96 (0.52)</td>
<td>Germany, continental anthropogenic pollution</td>
</tr>
<tr>
<td>Mamouri and Ansmann (2017)</td>
<td>0.23 (2.17)</td>
<td>0.41 (1.22)</td>
<td>1.41 (0.35)</td>
<td>Cyprus, continental anthropogenic pollution</td>
</tr>
<tr>
<td>Mamali et al. (2018)</td>
<td>0.14, 0.24 (3.57, 2.03)</td>
<td>0.19 (2.63)</td>
<td>0.60 (0.83)</td>
<td>Cyprus, fine non-dust aerosol fraction</td>
</tr>
<tr>
<td>This work</td>
<td>0.12 (4.17)</td>
<td>0.19 (2.63)</td>
<td>0.60 (0.83)</td>
<td>Continental (clean to moderately polluted)</td>
</tr>
</tbody>
</table>

Notes:
- MEE = $\alpha_a/(V_a \cdot \rho_a)$
- $c_v = V_a/\alpha_a$
Table 6. Main characteristics of the dataset of the EARLINET continental sites considered in this study. The listed dataset refers to the data downloaded from the EARLINET site (last access on the 11th of January 2018).

<table>
<thead>
<tr>
<th>Station</th>
<th>Number of points at 355 and at 532 nm)</th>
<th>Altitude range (Δz, in km)</th>
<th>Period</th>
</tr>
</thead>
<tbody>
<tr>
<td>40.33 N, 18.10 E, 30 m a.s.l.</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>51.35 N, 12.43 E, 90 m a.s.l.</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Potenza (PO)</strong></td>
<td>1244 – 219</td>
<td>1.5 – 4</td>
<td>May2000 – Aug2009</td>
</tr>
<tr>
<td>40.6 N, 15.72 E, 760 m a.s.l.</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Hamburg (HH)</strong></td>
<td>243 – n.a.</td>
<td>0.5 – 4</td>
<td>Apr2001 – Oct2002</td>
</tr>
<tr>
<td>53.57 N, 9.97 E, 25 m a.s.l.</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Madrid (MA)</strong></td>
<td>n.a. – 492</td>
<td>0.5 – 4</td>
<td>Jun2006 – Jun2008</td>
</tr>
<tr>
<td>40.45 N, 3.73 E, 669 m a.s.l.</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 7 Mean LR discrepancies between our model results and EARLINET measurements and weighted LR at 355 and 532 nm for the considered EARLINET stations.

<table>
<thead>
<tr>
<th>Station</th>
<th>( \lambda = 355 \text{ nm} )</th>
<th>( \lambda = 532 \text{ nm} )</th>
<th>( \lambda = 355 \text{ nm} )</th>
<th>( \lambda = 532 \text{ nm} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>LC</td>
<td>10</td>
<td>18</td>
<td>51.8</td>
<td>44.5</td>
</tr>
<tr>
<td>LE</td>
<td>6</td>
<td>9</td>
<td>52.6</td>
<td>51.0</td>
</tr>
<tr>
<td>PO</td>
<td>17</td>
<td>7</td>
<td>44.9</td>
<td>57.2</td>
</tr>
<tr>
<td>HH</td>
<td>5</td>
<td>-</td>
<td>53.3</td>
<td>-</td>
</tr>
<tr>
<td>MA</td>
<td>-</td>
<td>6</td>
<td>-</td>
<td>54.2</td>
</tr>
</tbody>
</table>
Table 8. Main characteristics of the ALC and co-located sun-sky radiometer equipment located at the considered ALICENET sites.

<table>
<thead>
<tr>
<th>Site type</th>
<th>ALC model</th>
<th>ALC firmware</th>
<th>Sun photometer model</th>
</tr>
</thead>
<tbody>
<tr>
<td>ASC</td>
<td>alpine</td>
<td>Nimbus CHM150104</td>
<td>0.743</td>
</tr>
<tr>
<td>SPC</td>
<td>rural</td>
<td>Nimbus CHM110115</td>
<td>0.556</td>
</tr>
<tr>
<td>RTV</td>
<td>semi-rural</td>
<td>Nimbus CHM070052</td>
<td>0.720</td>
</tr>
</tbody>
</table>
Table 9. Results of the comparison between the AOT measured by sun-photometers and the one derived by ALCs (model-based and fixed LR inversion schemes) at three ALICENET stations. Mean differences (expressed in terms of \(\langle d\text{AOT} \rangle = \langle (\text{AOT}_{\text{ceil}} - \text{AOT}_{\text{phot}}) \rangle\), \(\langle |d\text{AOT}| \rangle\), \(\langle d\text{AOT}/\text{AOT} \rangle\) and \(\langle |d\text{AOTI}/\text{AOT}| \rangle\)) are reported with their standard deviations.

| ALICENET sites | \(\langle d\text{AOT} \rangle\) | \(\langle |d\text{AOT}| \rangle\) | \(\langle d\text{AOT}/\text{AOT} \rangle\) | \(\langle |d\text{AOTI}/\text{AOT}| \rangle\) |
|----------------|----------------|----------------|----------------|----------------|
| ASC            |                |                |                |                |
| LR = 52 sr     | \(-0.004 \pm 0.015\) | \(0.010 \pm 0.013\) | \(-0.25 \pm 0.57\) | \(0.31 \pm 0.35\) |
| LR = 38 sr     | \(0.002 \pm 0.021\) | \(0.009 \pm 0.015\) | \(0.31 \pm 0.58\) | \(0.33 \pm 0.35\) |
| SPC            |                |                |                |                |
| LR = 52 sr     | \(-0.001 \pm 0.020\) | \(0.013 \pm 0.016\) | \(-0.005 \pm 0.28\) | \(0.19 \pm 0.20\) |
| LR = 38 sr     | \(0.021 \pm 0.026\) | \(0.026 \pm 0.02\) | \(0.33 \pm 0.35\) | \(0.38 \pm 0.26\) |
| RTV            |                |                |                |                |
| LR = 52 sr     | \(-0.003 \pm 0.019\) | \(0.011 \pm 0.014\) | \(-0.043 \pm 0.24\) | \(0.16 \pm 0.18\) |
| LR = 38 sr     | \(0.004 \pm 0.020\) | \(0.014 \pm 0.014\) | \(0.11 \pm 0.49\) | \(0.33 \pm 0.30\) |
|                | \(0.016 \pm 0.023\) | \(0.021 \pm 0.018\) | \(0.44 \pm 0.59\) | \(0.49 \pm 0.45\) |
|                | \(0.003 \pm 0.019\) | \(0.013 \pm 0.013\) | \(0.088 \pm 0.460\) | \(0.31 \pm 0.27\) |
Figure 1. Schematic of the two-step model structure developed to obtain, as a result, functional relationships between the aerosol backscatter (βₐ) and the aerosol extinction, surface area and volume (αₐ, Sₐ and Vₐ, respectively).
Figure 2. Scatterplots of a) $\alpha$ (km$^{-1}$), b) $S_a$ (cm$^3$/cm$^3$) and c) $V_a$ (cm$^3$/cm$^3$) vs backscatter $\beta_a$ (km$^{-1}$ sr$^{-1}$) and relevant relative errors (panels d, e, f, respectively) as derived from 20000 model computations (blue points) at $\lambda = 1064$ nm. Red dots and error bars are the average values per decade of $\beta$ and their standard deviations, green lines are the 7th-order polynomial fit curve of the 20000 points.
Figure 3. Upper plots: scatterplots of LR (sr) versus $\beta_a$ (km$^{-1}$ sr$^{-1}$) at: a) 355 nm; b) 532 nm; c) 1064 nm (blue points). The 7th-order polynomial fit curve (green lines) and the average values per decade of $\beta$ together with their standard deviations (red points and red vertical bars, respectively) are also reported. Horizontal black lines are mean values of the ‘weighted-LR’ and ± 1 s. d. (solid and dotted lines, respectively). Lower plots: relative errors associated with the model-derived LR at d) 355 nm; e) 532 nm; f) 1064 nm.
Figure 4. Scatterplots of LR (sr) versus $\beta_a$ (km$^{-1}$ sr$^{-1}$) at 355 nm as simulated by our model (colored region) and measured by EARLINET lidars (black open circles) in Hamburg (Germany) (a), Lecce (Italy) (b), Leipzig (Germany) c) and Potenza (Italy) (d). The color area is the region of simulated values, the color code indicating the number of simulated values in each $\beta_a$-LR pair (see legend). In particular, the color-2D histogram is computed using a semi-logarithmic box consisting of 10 equally spaced bins per decade of $\beta_a$ in the x-axis and 5 spaced LR values in the y-axis.
Figure 5. Model-simulated (blue) and lidar measured (red) LR vs $\beta_a$ mean curves at 355 nm calculated per 10 equally spaced bins per decade of $\beta_a$ in (a) Hamburg, (b) Lecce, (c) Leipzig, and (d) Potenza EARLINET lidar station. Vertical bars are the associated standard deviations.
Figure 6. Time-height cross-section of the aerosol extinction coefficients $\alpha_a$ [km$^{-1}$], as derived at 1064 nm on 26 June 2016 by the ALICENET ALC of Aosta San Christophe (Northern Italy). The orange circle points and the pink line are the AOT values (right y-axis, panel b) measured by a co-located POM-02L radiometer and estimated from the ALC following our approach.
Figure 7. a) geographical map of the ALC network ALICENET. The red circles highlight the selected sites for this study: Aosta San Christophe (ASC), San Pietro Capofiume (SPC) and Rome Tor Vergata (RTV). b-d) Histograms of the differences between the hourly-mean coincident AOTs at 1064 nm as derived by ALCs and measured by photometers, at ASC, SPC and RTV, respectively. The different colors (red, blue and black) depict the different inversion schemes: model-based inversion scheme, LR = 38 sr and LR = 52 sr, respectively. In each panel the values of the average measured AOT (and its associated standard deviation) and of the number of considered pairs are also reported.
Figure 8. Time-height cross-section of the aerosol volume concentration at Aosta San Christophe for 29 December 2016 (upper panel) and 05 September 2017 (lower panel). The right y-axis reports the volume concentration measured at surface through TSI and Fidas®200s OPCs (upper and lower panels, grey curves) and the ALC-derived volume concentration at 75
m (pink curves). The grey crosses and the pink triangles refer to the daily mean aerosol volume value derived by OPCs and ALC measurements, respectively. The horizontal bars in the upper part of the panels indicate the ranges (RH<60%, 60%<RH<90% and RH>90%, respectively) of the measured in-situ RH during the analyzed days.
Figure 9. Daily-resolved aerosol mass concentration at SPC, for the period June – July 2012, estimated from ALC-derived aerosol volume data at 225 m a.s.l. converted into mass using a fixed particle density $\rho_a = 2 \mu g/m^3$ (blue dotted line) and a variable $\rho_a$ between 1.5 - 2.5 $\mu g/m^3$ (shaded blue area). The red solid line is the daily PM10 concentration as measured by the local Air Quality agency (ARPA). Vertical yellow shaded stripes indicate the presence of dust events.
Appendix A: Model sensitivity tests

To evaluate the proposed continental model configuration (hereafter CM0) and discuss its sensitivity to the variability of the employed parameters, an overview of the impact on the model results produced by changing the limit of the variability ranges of these parameters (i.e. using different model configuration, CMX) is given in this section.

The varied model (CMX-CM0) mean difference on the considered optical property (OP) has been quantified through the following equation:

\[
\frac{\Delta \text{OP}}{\text{OP}} = \left( \frac{1}{{N_{\text{bin}}}} \right) \sum_{i=1}^{N_{\text{bin}}} \left[ \langle <\text{OP}_\text{CMX},i \rangle - \langle <\text{OP}_\text{CM0},i \rangle \rangle \right] / \langle <\text{OP}_\text{CM0},i \rangle \rangle,
\]

where \(N_{\text{bin}}\) is the total number of defined bins of \(\beta_a\).

The results of the mean differences of \(\alpha_a\) and LR for different ranges of \(\beta_a\) and for the whole \(\beta_a\) interval are reported on table A1, where relevant sensitivity cases (i.e. relative mean difference greater than 1%) at \(\lambda=355\) nm have been taken into account.

CM1 refers to a model configuration without the first aerosol mode (\(N_{1,2}=0\)). The overall decrease on the values of \(\alpha_a\) and LR (around 3-4%) is due to the sum of significant and opposite effects for low and high values of \(\beta_a\) where \(<\Delta \alpha_a/\alpha_a>\) and \(<\Delta \text{LR}/\text{LR}>\) are of the order of -6% and 8%, respectively. Removing the coarser aerosol mode (\(N_{1,2}=0\)), causes positive mean values for \(<\Delta \alpha_a/\alpha_a>\) and \(<\Delta \text{LR}/\text{LR}>\) of the order of 5% (sensitivity case CM2). In this case, the largest impact is observed for the \(\beta_a\) range between \(2 \times 10^{-3}\) and \(2 \times 10^{-1}\) km\(^{-1}\)sr\(^{-1}\).

An opposite result is obtained by decreasing the upper bound of the \(\tau_2\) variability range (\(\tau_2=0.03 - 0.05\) \(\mu\)m, CM3). In fact also this model configuration leads to lower \(\alpha_a\) and LR (\(<\Delta \alpha_a/\alpha_a>\) and \(<\Delta \text{LR}/\text{LR}>\) are equal to -6%, approximately). In this case, the variation on the \(\tau_2\) parameter affects the higher ranges of \(\beta_a\) (\(\beta_a=2 \times 10^{-4}-2 \times 10^{-2}\) km\(^{-1}\)sr\(^{-1}\)). Higher modal radii for the coarse-mode particle (\(\tau=1 - 1.2\) \(\mu\)m) in CM4 configuration leads to the increase of the contribution of model-generated points with higher \(\beta_a\) and causes lower values of \(\alpha_a\) and LR (\(<\Delta \alpha_a/\alpha_a>\) and \(<\Delta \text{LR}/\text{LR}>\) are equal to -5%, approximately) only for high values of \(\beta_a\) (\(\beta_a=2 \times 10^{-2}-2 \times 10^{-1}\) km\(^{-1}\)sr\(^{-1}\)), whereas the effect over the whole \(\beta_a\) range is around -1%.

The CM5 configuration accounts for the presence of more absorbing particles in the first aerosol mode, where the lower bound of \(m_{1,im}\) has been increased by a factor of 10 (\(m_{1,im}=0.1-0.47\)). This produces a significant effect only for the lower values of \(\alpha_a\) (\(\beta_a=2 \times 10^{-3}-2 \times 10^{-2}\) km\(^{-1}\)sr\(^{-1}\)), with an increase of \(\alpha_a\) and LR of approximately 4%. On the contrary, increasing the lower bound of the real part of the second aerosol mode refractive index (\(m_{1,2}=1.55-1.70\)) has a large impact on the considered parameters. In fact, the CM6 configuration largely underestimates both \(\alpha_a\) and LR (around -15% for both parameters) for all \(\beta_a\) ranges.

The CM7 configuration refers to the impact of the total number of particles at the ground (\(N_{\text{tot}}\)). In this case, decreasing the upper bound of the variability range of \(N_{\text{tot}}\) by a factor of 2 (\(N_{\text{tot}}=500-5000\) cm\(^{-3}\)) lowers the mean values of \(\alpha_a\) and LR of around 5%. Nevertheless, this effect is totally due to the contribution of the \(\beta_a\) values between \(2 \times 10^{-3}\) and \(2 \times 10^{-2}\) km\(^{-1}\)sr\(^{-1}\), where \(<\Delta \alpha_a/\alpha_a>\) and \(<\Delta \text{LR}/\text{LR}>\) are around -10%. Assuming no increase with altitude for \(\sigma_{1,2}\) (sensitivity case CM8) produces relevant differences on the mean values of \(\alpha_a\) and LR. In CM8, the overall overestimation of these two parameters is quite limited (\(<\Delta \alpha_a/\alpha_a> = 6.3\) and \(<\Delta \text{LR}/\text{LR}> = 6.4\)), whereas a large and opposite impact is observed for
low and high values of $\beta_a$. In fact, $<\Delta\alpha/\alpha_a>$ ($<\Delta LR/LR>$) is equal to -14.1 (-13.9) and 18.5 (19.0) for $\beta_a = 2 \times 10^{-5} - 2 \times 10^{-4}$ and $\beta_a = 2 \times 10^{-3} - 2 \times 10^{-4}$ km$^{-1}$sr$^{-1}$, respectively. As explained by Barnaba et al. (2007), the dependence of $\sigma_{1,2}$ to the altitude can be associated to the fact that, when increasing the distance from the main aerosol sources, the particle processing is more efficient.

Table B1. Mean differences of $\alpha_a$ and LR between different model sensitivity cases and the proposed continental model configuration.

<table>
<thead>
<tr>
<th>Model configuration</th>
<th>$\beta_a$(km$^{-1}$sr$^{-1}$) $2\times10^{-5}$ - $2\times10^{-4}$</th>
<th>$\beta_a$(km$^{-1}$sr$^{-1}$) $2\times10^{-3}$ - $2\times10^{-2}$</th>
<th>$\beta_a$(km$^{-1}$sr$^{-1}$) $2\times10^{-2}$ - $2\times10^{-1}$</th>
<th>$\beta_a$(km$^{-1}$sr$^{-1}$) $2\times10^{-1}$ - $2\times10^{0}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>CM1 (N$_1$%=0)</td>
<td>$&lt;\Delta\alpha/\alpha_a&gt;$ (%) $&lt;\Delta LR/LR&gt;$ (%)</td>
<td>$&lt;\Delta\alpha/\alpha_a&gt;$ (%) $&lt;\Delta LR/LR&gt;$ (%)</td>
<td>$&lt;\Delta\alpha/\alpha_a&gt;$ (%) $&lt;\Delta LR/LR&gt;$ (%)</td>
<td>$&lt;\Delta\alpha/\alpha_a&gt;$ (%) $&lt;\Delta LR/LR&gt;$ (%)</td>
</tr>
<tr>
<td>CM2 (N$_3$%=0)</td>
<td>4.7 4.9 8.6 8.9 2.8 2.7 5.3 5.4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CM3 ($r_a=0.03 - 0.05$ µm)</td>
<td>-2.0 -1.7 -10.3 -10.2 -8.9 -8.2 -6.7 -6.4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CM4 ($r_a=1.0 - 1.2$ µm)</td>
<td>&lt;1 &lt;1 -2.1 -2.0 -5.24 -5.3 -1.2 -1.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CM5 (m$_{110}=0.1-0.47$)</td>
<td>4.3 4.2 &lt;1 &lt;1 &lt;1 &lt;1 1.8 1.8</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CM6 ($m_a=1.55 - 1.70$)</td>
<td>-10.9 -10.9 -16.2 -16.3 -18.9 -19.1 -15.3 -15.3</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CM7 (N$_{100}=500-5000$)</td>
<td>&lt;1 &lt;1 &lt;1 &lt;1 &lt;1 11.2 -10.7 3.7</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CM8 ($\sigma_1, \sigma_2$ constant)</td>
<td>-14.1 -13.9 6.4 6.1 18.5 19.0 6.3 6.4</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Appendix B: Model-based functional relationships at 355 and 532 nm

The parameters of the seventh-order polynomial fit used to derive the functional relationships between \( \log(x) \) and \( \log(y) \) (where \( x = \beta_a \) and \( y = \alpha_a, S_a \) or \( V_a \)) at \( \lambda = 355 \) and 532 nm are reported in Tab. A1 and Tab. A2, respectively.

Table A1. Parameters of the Seventh-Order Polynomial Fits \( y = a_0 + a_1x + a_2x^2 + a_3x^3 + a_4x^4 + a_5x^5 + a_6x^6 + a_7x^7 \) for \( \lambda = 355 \) nm, with \( x = \log(\beta_a) \) (in km\(^{-1}\) sr\(^{-1}\) unit) and \( y = \log(\alpha_a, S_a \) or \( V_a \) in (km\(^{-1}\)), cm\(^2\)/cm\(^3\) and cm\(^3\)/cm\(^3\), respectively).

<table>
<thead>
<tr>
<th>Functional relationship at 355 nm</th>
<th>Extinction coefficient</th>
<th>Surface area (km(^{-1}))</th>
<th>Volume (cm(^2)/cm(^3) and cm(^3)/cm(^3), respectively)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( a_0 )</td>
<td>3.797837507651898</td>
<td>12.019452592845141</td>
<td>-5.314834128998254</td>
</tr>
<tr>
<td>( a_1 )</td>
<td>3.294032541389781</td>
<td>30.825966279368547</td>
<td>2.500484347793244</td>
</tr>
<tr>
<td>( a_2 )</td>
<td>0.962603336867675</td>
<td>24.518531616019207</td>
<td>-1.196109537503000</td>
</tr>
<tr>
<td>( a_3 )</td>
<td>0.241796629870675</td>
<td>10.625241994796593</td>
<td>-1.583236058579546</td>
</tr>
<tr>
<td>( a_4 )</td>
<td>0.064609145804688</td>
<td>2.634051072085453</td>
<td>-0.681801883947768</td>
</tr>
<tr>
<td>( a_5 )</td>
<td>0.017721752150233</td>
<td>0.373150843707711</td>
<td>-0.145232662646142</td>
</tr>
<tr>
<td>( a_6 )</td>
<td>0.002722551625862</td>
<td>0.027971628176431</td>
<td>-0.015471229968392</td>
</tr>
<tr>
<td>( a_7 )</td>
<td>0.000157245409783</td>
<td>0.000854381337164</td>
<td>-0.000658925756875</td>
</tr>
</tbody>
</table>
Table A2. Parameters of the Seventh-Order Polynomial Fits \( y = a_0 + a_1x + a_2x^2 + a_3x^3 + a_4x^4 + a_5x^5 + a_6x^6 + a_7x^7 \) for \( \lambda = 532 \) nm, with \( x = \log(b_a) \) (in km\(^{-1}\) sr\(^{-1}\) unit) and \( y = \log(a_a, S_a, or V_a) \) in (km\(^{-1}\), cm\(^3\)/cm\(^3\) and cm\(^3\)/cm\(^3\), respectively).

<table>
<thead>
<tr>
<th>Functional relationship at 532 nm</th>
<th>Extinction coefficient</th>
<th>Surface area</th>
<th>Volume</th>
</tr>
</thead>
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<tr>
<td>( a_0 )</td>
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<td>-0.000658925736875</td>
</tr>
</tbody>
</table>
Appendix C: Model – EARLINET comparison at 532 nm

Figure C1 depicts the result of the comparison between EARLINET stations and our developed model (red and blue curves, respectively) in terms of ‘mean’ LR per bin of $\beta_a$ at $\lambda=532$ nm. Note that only $\beta_a$ bins containing at least 1% of the total modeled data were considered. Similarly to the results at 355 nm shown in section 4.1, a general good agreement between the modeled and the measured LR values is found. As attested by the low value of the mean discrepancy of Table 6, the modeled curve well fits with Madrid observations. Some major deviations are found for Lecce, which, however, at 532 nm, has a very low number of considered points (i.e. 109).

Figure C1. Model-simulated (blue) and lidar measured (red) LR vs $\beta_a$ mean curves at 532 nm calculated per 10 equally spaced bins per decade of $\beta_a$ in a) Madrid, b) Lecce, c) Leipzig, and d) Potenza EARLINET lidar station. Vertical bars are the associated standard deviations.
Appendix D: Model sensitivity tests for optimal configurations at LC and PO sites

According to the results reported in Tab. B1, two model configurations (CM0a and CM0b) have been set up to better reproduce the EARLINET observations of LR vs $\beta_a$ at LC and PO sites, respectively. The comparison between these two configurations, the EARLINET measurements and the CM0 set-up are illustrated in Fig. B1 (panel a and b for LC and PO, respectively) in terms of LR mean value curves per 10 equally spaced bins per decade of $\beta_a$. Blue and red colors have the same meaning of Fig. 5 (i.e. CM0 model and observation curves, respectively), black curves refer to the LR vs $\beta_a$ estimated through the CM0a and CM0b model versions for LC and PO stations, respectively. Vertical bars are the associated standard deviations.

The only difference between CM0a and CM0 configuration consists in the upper bound of the variability range of $N_{\text{tot}}$ (5000 vs 10000 cm$^{-3}$ at ground, respectively). This modification seems to fit the observed LR vs $\beta_a$ behavior at 355 nm. The upper bound $N_{\text{tot}}$ value is similar to the one (i.e. $N_{\text{tot}}$ upper bound =3000 cm$^{-3}$ at ground) used in the work of Barnaba et al. (2007) to characterize the optical properties of the continental aerosol present over southeastern Italy. The computed mean model-measurement LR relative difference between CM0a configuration and LC Earlinet measurements is around 5%.

Similarly, the CM0b configuration uses the same value for the upper bound of $N_{\text{tot}}$ variability range and, in addition, higher values of the $r_1$ variability range of (1.0 – 1.2 $\mu$m vs 0.3 - 0.5 $\mu$m, respectively). As highlighted by the panel b of Fig. B1, this model configuration allows well reproducing the LR vs $\beta_a$ behavior derived by EARLINET lidar Raman measurements at 355 nm. This result seems to indicate the presence of coarser aerosols in a clean continental environment. In comparison to the CM0 model, the mean model-measurement LR relative difference decreases from 17% to 6%.

![Figure D1. Model-simulated (blue and black lines) and lidar measured (red lines) LR vs $\beta_a$ mean curves at 355 nm calculated per 10 equally spaced bins per decade of $\beta_a$ for the LC and PO EARLINET lidar stations (panel a and b, respectively). Blue color refers to CM0 model configuration, black color to CM0a and CM0b model configurations adapted to LC and PO sites, respectively.](image-url)
Appendix E: ALC vs sun-photometer AOTs

To have sense of both absolute and relative errors of AOT, we reported in this section the scatter plots between the hourly-mean coincident AOTs at 1064 nm as derived by ALC model-based approach and those measured at 1020 nm by the sun-photometers installed at RTV, SPC and ASC, respectively (Figure E1, E2 and E3). The corresponding linear fit $y = bx$ (red line), where $x =$ sun-photometer AOT, $y =$ Nimbus CHM15k AOT are also shown in the plots. The values of the correlation coefficients for the three sites ($R = 0.77$, $R=0.72$ and $R=0.73$ for RTV, SPC and ASC, respectively) attest a relatively good agreement between the two AOT measurements.
Figure E2. Scatter plot between the hourly-mean coincident AOTs at 1064 nm as derived by the ALC model-based approach and measured at 1020 nm by the SKYRAD photometer at SPC. The red line represents the linear fit $y = bx$ between the two datasets, where $x =$ sun-photometer AOT; $y =$ Nimbus CHM15k AOT.
Figure E3. Scatter plot between the hourly-mean coincident AOTs at 1064 nm as derived by the ALC model-based approach and measured at 1020 nm by the SKYRAD photometer at ASC. The red line represents the linear fit $y = bx$ between the two datasets, where $x =$ sun-photometer AOT; $y =$ Nimbus CHM15k AOT.