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Atmospheric Measurement Techniques
Discussions

Interactive comment on "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" *by* Davide Dionisi et al.

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Reply to referee#3

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. Il 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 (mi), with both its real and imaginary component (mi = mr_i - i × mim_i). The Mie theory for spherical particles of radius ri and refractive index mi are then used to compute the extinction and backscatter coefficients (see equations below).The Mie theory for spherical particles of radius ri and refractive index mi (= mr_i - i × mim_i) is then used to compute the extinction and backscatter coefficients of mode i (see equations 7-8)’.

#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.Hereafter, for the sake of simplicity, we will report, as an example of the refractive index variability of each mode, only the values obtained at λ=355 nm. The values of mi at λ=532 and 1064 nm are reported 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.

#12Line 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 ßa at λ=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 λ=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 (line299): ‘For the EARLINET Raman stations fulfilling these requirements, Figure 4 depicts the results of the model-measurements comparison in terms of LR vs ßa at λ=355 nm (the corresponding results at λ=532 nm, including Madrid in place of Hamburg, are given in Appendix C, Figure C1)’The corresponding results at λ=532 nm for those system having this green channel are given in Appendix C, Figure C1’.

#15 Line 313: ãËŸA Ëİ ULRãËŸA ° U_mod,ãËŸA Ëİ ULRãËŸAU° _meas 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 z2*P(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 with the equations are in the pdf supplement file.

#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 at 1020 nm 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 attached at the end of the file and their captions are, respectively:

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 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 AERONET 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 AERONET 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 Va and ßa at λ = 1064 nm (see Table 3 and Figure 2c) to retrieve aerosol volume profiles from ALC-derived- ßa 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 Va 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 Va 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 Va 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'.The impacts of aerosol size distribution in the retrieval of PM2.5 using ceilometers and the relationship between relative humidity and PM2.5/ceilometer-backscatter ratio have been discussed in several studies (e.g. Li et al., 2016, Li et al., 2017). These impacts are important in our volume estimates and relevant errors (e.g. also Barnaba et al., 2010; Adam et al., 2012). However, the quantitative characterization of the impact of RH and of particle number concentration to the ALC retrieved volume is beyond the aims of this paper and would require a full paper itself. This theme is also discussed in Diemoz et al. 2018 (being submitted to this same issue).

#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: a = 2 g/cm3 , with a range between 1.5-2.5 g/cm3). We took the opportunity of this comment to specify that: 'This range covers approximately the mean 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 ± 6.5 g/cm3, <|dPM10|> = 5.2 ± 4.7 g/cm3 and <(dPM10/PM10)> = 0.15 ± 0.27.This is confirmed by the good agreement between the ALC-derived and the ARPA PM10 values (Fig. 9) with a correlation coefficient (R) of 0.73. In fact, mean, absolute mean and relative differences, between the two series are: <dPM10> = 2.8 ± 6.5 μg/m3, <|dPM10|> = 5.2 ± 4.7 μg/m3 and <(dPM10/PM10)> = 0.15 ± 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 (ßa, αa) and physical (Sa, Va) properties (with associated uncertainties and limitations) by means of relatively simple and robust instruments.Overall, the main advantage of the proposed approach is the possibility to operationally (i.e. h24/7) retrieve fairly reliable, remote sensing profiles of aerosol optical (ßa, αa) and physical (Sa, Va) properties (with associated uncertainties and limitations) by means of relatively simple and robust instruments. Conversely, we know accurate measuring of aerosol optical properties to require rather expensive, in situ instruments. Furthermore, measurements of aerosol volume and surface area represent a rather difficult task even for in-situ, ground alone observations.

#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).This approach can represent a valid option to extend the capabilities of ALCs at characterizing the aerosol vertical distribution, providing important information 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).

Please also note the supplement to this comment:
https://www.atmos-meas-tech-discuss.net/amt-2018-79/amt-2018-79-AC3-supplement.pdf

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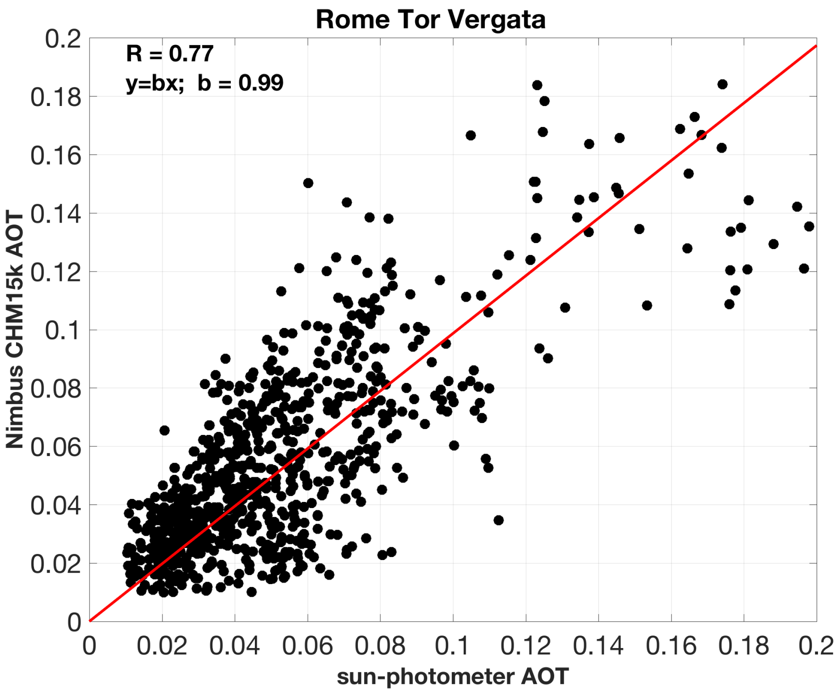
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Fig. 1.

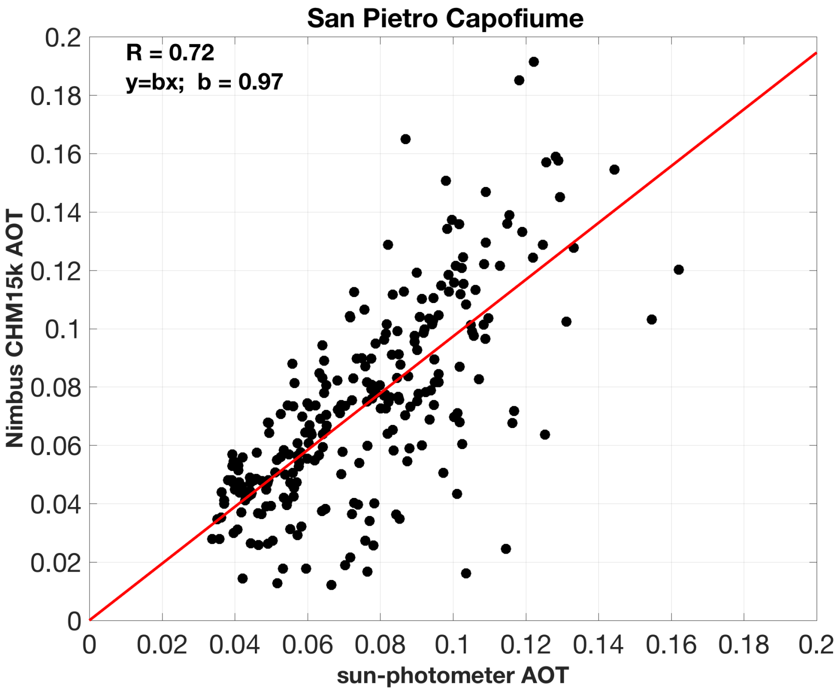
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Fig. 2.

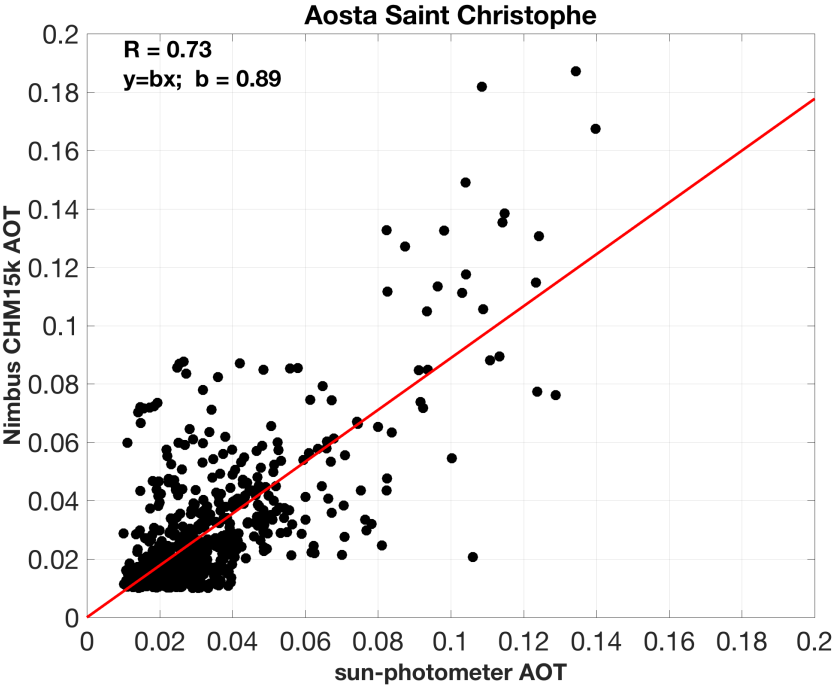
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Fig. 3.