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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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#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.

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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 per-

formed for empirical ranges of particle sizes and refractive indices, yielding conversion factors which are stored into a look-up table. As the influence of unsperical 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 re-

sults 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 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 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

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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 Âż rmi_0, i.e. for a large aequous droplet. Replace by mRh = mW + : : :

- Corrected, thank you for this remark

#4 Line 187: In eq. 4 and 5, rim_RH and miRH are the: : : should be : : :rmi_RH: : :

- Corrected, thank you for this remark

#5 Line 225: It is unclear to me what that means – what is 1%?

- We meant the βa region where each of the 10 equally-spaced bins per decade of βa contains at least 1% of the simulated points for the various aerosol parameters (i.e. αa, Sa and Va). We then reformulated the sentence: 'The red vertical bars of Figure 2 also highlight the ranges of αa, Sa and Va which are statistically significant,

i.e. those in which, at λ = 1064 nm, the model provides at least 1% of the total points per corresponding bin of ßa.'

#6 L 235: weighted-LR' _ 1 s. d. -> write: standard deviation

- Corrected, thank you for this remark

#7 Line 305 and Fig 4: 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. . .'

#8 Line 319ff: 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 βa (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).

#9 Line 420: 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).

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