Anonymous Referee #2

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We would like to thank the reviewer for the thorough revision of the manuscript and for suggestions! We hope we addressed the comments accordingly.

<u>We use across the text (below) the following highlights</u>:

In red, reviewer's comments.

In black, our comments.

In green, the citations from the manuscript submitted.

In blue, the changed text for the revised manuscript.

Before answering to the reviewer's questions/comments etc, we would like to indicate a few changes that we have done to the initial manuscript. Those changes are not related with the data processing and analysing or its scientific content but rather with text cosmetics.

- Based on suggestions from Earlinet community, we changed the stations acronyms from two letters to three letters, according to the new nomenclature. This was already implemented in Part II. Thus, all the figures with references at station acronyms (on labels or title) were changed for clarity and consistency. Similar holds for tables.

- based on editor's suggestion for Part II, we changed N to North (for N America and N Africa)

-based on suggestions from the editor of Part II, we refer to the stations in the text by the full name instead of using acronyms.

- EAE355/532 was changed to EAE in the text (two instances: pp 11, line 18 and line34).

- a list with the acronyms used in the text was added in Supplement (as in Part II) for consistency. The reference at the acronyms list is mentioned at the end of Introduction.

A list of acronyms used in the current work is given in the Supplement (Table S1).

- figs. 10, 11, 12, 13 and 14 (now 7, 8, 9, 10 and 11): we added a), b) etc for each plot for an easier reference. We made small changes to the text and changed the3 figures caption:

Fig. 7 upper right plot -> Fig. 7 a

In Fig. 7, the first two left plots we show -> in Fig. 7 a-b we show

Fig. 7 lower left plots -> Fig. 7 c-d

Figure 7 caption:

Fig. 10. Measurements with the same source at Thessaloniki ('th) and Bucharest ('bu'). Event: 20140909-20140910. Left plots: (first) fires location seen during back-trajectories from each station (colour coded), (second) histogram of the fires occurrence in each geographical grid, (third) longitude and latitude of the fires' location versus fires' occurrence time, (forth) longitude and latitude of the fires' location versus measurement time at the two locations. Right plots: layers' altitude and intensive parameters for each station. Layers measuring the common fire are shown by arrows. The geographical location of the common fire is shown on histogram by an arrow.

Figure 7. Measurements with the same source in Thessaloniki ("the") and Bucharest ("ino") during 20140909-20140910. a) Fires location seen during back-trajectories from each station (colour coded); b) Histogram of the fires occurrence in each geographical grid; c) Longitude and latitude of the fires' location versus fires' occurrence time; d) Longitude and latitude of the fires' location versus measurement time at the two locations; e–i) layers altitude and intensive parameters for each station. Layers measuring the common fire are shown by arrows. The geographical location of the common fire is shown on histogram by an arrow.

Figure 8, left -> Figure 8 a)

Fig. 8, right -> Fig. 8 b)

See lower plot of Fig. 7 -> See Fig. 8 b)

Figure 8 caption:

Fig. 11. Backtrajectories and location of fires along backtrajectories within 100 km and +/- 1h for Bucharest ('bu') on 20140909 (left) and Thessaloniki ('th') on 20140910 (right). Lower plots show the altitude (a.s.l.) of the backtrajectories function of time. The fires' time is shown as well (see arrow location). The blue square denotes the geographical location of the common fire. See text for more details.

Figure 8. Backtrajectories and location of fires along backtrajectories within 100 km and +/- 1h for Bucharest ("ino") on 20140909 (a) and Thessaloniki ("the") on 20140910 (b). Lower plots show the altitude (a.s.l.) of the backtrajectories versus time. The common fire location is marked with an arrow. See text for more details.

Fig. 9, upper right plot -> Fig. 9 e

blue arrows in Fig. 9 -> blue arrows in Fig. 9 (a-e)

green arrows in Fig. 9 -> green arrows in Fig. 9 (a-e)

magenta arrows in Fig. 9 -> magenta arrows in Fig. 9 (a-e)

Figure 9 caption:

Fig. 12. LRT as measured at "at" on 20170713. First two BB layers are considered "mixed" while the third "pure NA". The arrows show the location of the fires (left plot) and the location of the smoke layers (right plot).

Figure 9. LRT as measured over Athens ("atz") on 20170713. a) Location of the fires; b) Histogram of the fires. The North America fires are marked by arrows; c) Fires' coordinates versus fires' occurrence time; d) Fires' coordinates versus measurements time; e) Location of the layers marked by arrows. First two BB layers are considered "mixed" while the third (magenta) "pure NA"; f) – i) Intensive parameters.

The trajectory layer in Fig. 10 is the higher one (light blue). -> The trajectory layer in Fig. 10 c) is the higher one (light blue).

Figure 10 caption:

Fig. 13. Backtrajectories for layers shown in Fig. 12 for Athens ('at') station. First two layers are considered "mixed" while the third is considered "pure N America". See text for more explanation. The squares show the location of the fire.

Figure 10. Backtrajectories for layers shown in Fig. 9 for Athens ("atz") station. Layers in a) and b) are considered "mixed" while the layer in c) is considered "pure N America". See text for more explanation. The fire location is marked with an arrow.

Figure 11…. Upper plot shows the location of the fires…-> Figure 11… a) Location of the fires

Figure 11… The bottom plot shows the histogram of the fires -> Figure 11… b) Histogram of the fires

Figure 11 caption:

Fig. 14. SE Europe region formed by stations "at", "bu", "po", "sf" and "th". Upper plot shows the location of the fires detected by each station. Note that due to overlap some are not seen. The bottom plot shows the histogram of the fires detected by each station.

Figure 11. SE Europe region formed by stations Athens ("atz"), Bucharest ("ino"), Potenza ("pot"), Sofia ("sof") and Thessaloniki ("the"). a) Location of the fires detected by each station. Note that due to overlap some are not seen. b) Histogram of the fires detected by each station.

Figure 12 caption:

Fig. 15 Scatter plots between various two intensive parameters for SE region. The colour code of the points is station related (as labelled in the title). The colour code for the mean and STD values is related with the source origin (as stated on the plots).

Figure 12. Scatter plots between various two intensive parameters for SE region (LR@532 vs LR@355, LR@532 vs PDR@532, EAE355/532 vs BAE355/532, EAE355/532 vs BAE532/1064, EAE355/532 vs LR@532 and BAE532/1064 vs BAE355/532). The colour code of the points is station related (as labelled in the title). The colour code for the mean and STD values is related with the source origin (as stated on the plots).

Pp 4, line 18, pp 6, line 22: change 60 % to 65 %.

- Based on editor suggestion for part II, we do not discuss any more in Part II the other event for "common fire" analysis (here section 5.1). Thus, we will add few comments here at the end of section 5.1. Note that it was an error on the manuscript: we mistakenly wrote 20150602 instead of 20170602.

Initial:

„For the other event with common IP for the same source (20170529-20150602), the smoke was labelled as of 'single fire' as no other fires were identified along the backtrajectory. This event will be discussed in the subsequent paper."

Changed:

For the other event with common IP, recorded in Athens and Thessaloniki during 20170529-20170602, the smoke was labelled as of 'single fire' as no other fires were identified along the backtrajectory. The common fire occurred on 26th of May at midnight in Ukraine (48.171 N, 30.622 E) and it was recorded in Thessaloniki and Athens on 29/05 and 31/05 respectively. BAE@532/1064 value in Thessaloniki was less than half of that in Athens, while BAE@355/532 was larger for Thessaloniki. High BAE corresponds to higher backscatter at smaller wavelengths, which indicates a higher number of small particles. The values in Thessaloniki correspond to a higher number of small size particles (at 355 nm) and with a higher proportion of large particles (at 1064 nm) compared to the ones over Athens. CR_{BAE} (colour ratio of the backscatter Ångström exponents) increases from Thessaloniki to Athens (0.22 to 0.78, respectively), which suggests an increase with travel distance (time). As CR_{LR} (colour ratio of the lidar ratios) and EAE (extinction Ångström exponent) were not available to characterize the smoke in terms of age, we classified the smoke as aged based on the duration of the travel time.

Overall, we conclude that the number of common events as well as the number of the common IPs is limited and, thus, no thorough examination of these events is possible. The most important feature of this analysis is that it enables us to quantify the smoke as of 'single fire' or 'mixed' and hence explain various IP values. This kind of analysis can be successfully applied in the future, when more data become available.

<u>Answers to specific comments of the Referee:</u>

Significant part of the manuscript is dedicated to description of the procedure of data treatment. No question, it is important when large volume of data from different stations is analyzed. Still this is ACP, so may be it is better to put data treatment in Appendix? But this is up to the authors.

Thank you for this suggestion. We moved to Appendix Figs. 5, 6 and 8 as well as Table 2 (now they cite as Figs. S1, S2 and S4, Table S2). The figures and tables were re-numbered. We consider that Chapter 3 on data quality control is not large and thus we would like to keep it in the main manuscript. As different criteria involved in QC are discussed along various steps of the procedure, it is difficult to move sparse parts to Appendix. We moved to Appendix the description of the algorithm to determine the aerosol boundary layer (Section 3 in Supplement).

Data quality is important and Fig.4 probably should illustrate it. However, it rises a lot of questions.

Actually, every plot provides a lot of questions and reader will definitely be confused. The uncertainties should be provided to separate real results from artifacts.

Regarding the examples in Fig. 5 (not 4 as mentioned), we added the uncertainties as suggested. In general, we cannot comment precisely on the accuracy of the optical properties profiles as related to different factors. In the database of Earlinet such information is not provided and therefore we could not investigate how they originated (e.g. calibration region, depolarization constant etc). **The input data in the study were the b-files and e-files containing the optical parameters and associated errors (which were quality checked by Earlinet QC tools and approved by the PIs of the stations)**. However, a series of additional quality checks were implemented within our study (discussed in detail in the text). We performed an investigation about the profiles shown in Fig. 5 to show how we managed the IPs values.

For example, in Fig.4a, extinction. in upper layer (2000 m) at 532 nm is stable, but at 355 nm it oscillates. Is it real or just artifact?

Fig. 5a). In fact, this illustrated the special means of additional quality check that we conducted in our study. Note that large uncertainties were seen for backscatter at 532 nm above ~ 3km (x-axis not shown at full scale on left plot) and for extinction at 532 nm in the first range bins. Further, depolarization is shown as zero for few hundred meters, which is an artefact. We cannot say for sure if extinction at 532 nm has an artefact but likely 532 channel had some problems at this time. Therefore, we checked if the final IPs data set contains data from this measurement. There is no IP associated with this time stamp. The reason for this is that for these layers we did not detect any fire along backtrajectories and thus the data was dismissed (before any other quality checks).

On Fig.4b the peak of extinction is more narrow than that of backscattering. Why? At 3000 m depolarization at 355 nm becomes larger than at 532 nm. Is it real?

Fig. 5b). Extinction profiles for 355 and 532 are extracted from e-file (as mentioned in the text). For this particular case, there is 101 bins 'smooth running' (49 bins are used in b-files). Evaluation method is Raman in both b-files and e-files. Regarding the peaks, the extinction profiles may present artefacts towards last validated bins. The data were eliminated above 2.7 km (as provided in e-file). In the paper by Ortiz-Amezscua et al. (2017), the authors show profiles of backscatter and extinction as well as LR and PDR (their Fig. 8) for 00:00-01:00 interval. Their extinction profiles go up to ~3.5 km. Unfortunately, they do not report PDR@355nm. PDR@532 is ~ 3% (similar with the present plot, but note that their layer is estimated differently). We did not find a fire for this case either, so the data was eliminated from analyses. Most probably, extinction at 532 nm would have been eliminated when computing the mean values in the layer as we wouldn't have had 90 % of the data available. Looking at closer measurements in our dataset, we found the following PDR mean values in layers as: 23:28 on 08/07 PDR@355=2.96 and PDR@532=3.01, 06:29 on 09/07 PDR@355=2.84 and PDR@532=3.17, 15:22 on 09/07 PDR@355=2.77 and PDR@532=2.87. As seen, the values are very close in value. However, there are regions where PDR@355 > PDR@532. This is possible, and it is reported in literature e.g. Janicka et al. 2017, Harrig et al. 2019, Baars et al. 2019.

In Fig.4c,d when backscattering coefficients at355 nm are calculated, the reference points are not shown and it is not clear, if these exist.

Fig. 5c)-d). We did not investigate the reference points (nor showed on plots). The reference points are not mandatory variable in the Earlinet database and they are sporadically reported.

For **Fig. 5c)** the data processing was performed with in-house (PollyXT) algorithm and we have the following information: @355 'Ref.value 4 50 1/m*sr at 3500.5m', @532 'Ref.value 1 70 1/m*sr at 7352m', @1064 'Ref.value 6 0 1/m*sr at 8300.5m' (all using Photon Counting and Raman as evaluation method). We obtained the following IPs for the three layers (all with BB origin): 2.7 for BAE@355/532 (Ist layer), 2.6, 1.1 and 1.5 for BAE@532/1064, and 2.1, 5.6 and 4.1 % for PDR@532. Due to various criteria, the values for PDR@355 were not estimated.

For the plot in **Fig. 5d)** the following information is given in the file (processed with SCC): @b355 'find calibr. interval (width = 500m) between 5000 and 9000m with method: min. of sig ratio; bsc. ratio = 1.0E+000' (backscatter ratio method), @b532 'find calibr. interval (width = 500m) between 3000 and 9000m with method: min. of sig ratio; bsc. ratio = 1.2E+000' (backscatter ratio method), @b1064 'find calibr. interval (width = 500m) between 2000 and 5000m with method: min. of sig ratio; ' (iterative method). We obtained for both layers (with BB origin) the following IP values: 40 and 72 sr for LR@532, 2.3 and 0.9 for EAE, 1.7 and 1.8 for BAE@532/1064. As seen, the value of BAE@355/532 and LR@355 could not be estimated due to unreliable profile for backscatter at 355nm. However, we suspect that the backscatter profile at 532 nm is not accurate either (we do not know how ABR=1.2 was chosen).

Fig. 5e). Both layers were identified as having BB origin. The following IPs values were calculated: 20 and 31 sr for LR@355, 1.1 and 1.7 for EAE.

Fig. 5f). Only the second layer was identified as having BB origin. The following IPs were determined. LR@355=46sr, LR@532=91sr, EAE=0.25, BAE@355/532=1.9, BAE@532/1064=0.9, PDR@355=2.8%, PDR@532=2.2 %.

Fig. 5g). Only the first layer was identified as having BB origin. The following IPs were calculated: BAE@355/532=1.9, BAE@532/1064=1.3. Due to various criteria, PDR was not estimated.

Fig. 5h). This dataset was eliminated as considered to have no BB origin.

Fig. 5i). All layers have BB origin. The following IPs were calculated: 21sr for LR@355 (Ist layer), 67sr for LR@532 (Ist layer), 1.2 and -0.56 for EAE (Ist and IInd layer).

Note that all the profiles showing regions with PDR355 > PDR532 belong to the same event (long range transport from North America), recorded over 7-10 July 2013 in Warsaw, Belsk and Cabauw (discussed in part II). PDR is provided only by Warsaw ("waw") for this event and it is in accordance with results reported by Janicka et al. 2017. The PDR values retrieved for the entire three days period are small and very close in value, slightly larger for PDR355. Thus, for eight cases where we had estimates at both wavelengths, we obtained the mean and STD as: PDR355 = 2.5 ± 0.5 % and PDR532 = 2.4 ± 0.9 %.

We would like to mention again that it is hardly possible to thoroughly check each profile manually in full detail because of the very high number of profiles analysed (> 4000 profiles) and also to check how the individual retrievals were performed. We only have access at the final product (optical properties). It was behind the scope of this study to check how the raw data were processed with an in-house algorithm or SCC (to do that, firstly, raw data would have to be available; secondly, this kind of work is a study by itself and it needs huge amounts of resources). Conversely, we focused on post processing quality checks along various steps in the procedure. From these examples, we can see that many IPs were not fulfilling our QC criteria and thus rejected form analyses. We did not investigate in detail such situations when backscatter or extinction at 532 nm is larger than those at 355 nm or when PDR@355 > PDR@532. These situations are very rare but they can be real (e.g. Burton et al., 2015; Haarig et al., 2018; Hu et al., 2018; Stachlewska et al., 2018). Haarig et al. (2018) and Hu et al. (2018) report PDR355 > PDR532 for stratosphere while both PDR have larger values (~20%). For troposphere, Haarig et al. report PDR355 = 2 % ± 4 % and PDR532 = 3 % ± 2 %. Haarig et al. hypothesize that the missing coarse mode in the size distribution is responsible for the high spectral dependence of PDR. Stachlewska et al. (2018) record in a layer at 2.2 - 2.4 km PDR355 = 1.6 ± 0.2 and PDR532 = 0.3 ± 0.1, this being related to advection of smoke particles.

A tremendous work was put on QC of the data analysed in this study. We do not claim it is perfect but we believe that we have eliminate most of unreliable profiles. The purpose of the Table 3 was to show how different datasets were eliminated during various stages, following various criteria.

We add the following statement at the end of section 4.1, describing the Fig. S1.

…Most of the layers detected are situated between 1000 and 5000 m altitude (typically above PBL). However, the minimum layer bottom was found at 257.5 m while the highest layer top was found at 19,8 km. Minimum, maximum and the mean layer thickness were 300, 6862.5 and 1337.5 m. Please note that not all the layers shown here have BB origin (as this check is not performed yet).

The optical profiles shown in Fig. S1 illustrate the layer estimation and show also various questionable patterns for different optical variables. Our quality checks were meant to eliminate profiles (or parts of profiles) where suspicions in their high-quality arise. The examples shown in a), b) and h) were eliminated as they were considered of non-BB origin (as discussed later). For the profiles in c), all layers have BB origin. Various QC did not allow the estimation of the IPs based on non-reliable backscatter coefficient at 355 nm for second and third layer while PDR@355 was dismissed as well. For d) case, both layers have BB origin. QC did not allow the retrieval of various IPs based on non-reliable backscatter coefficient at 355 nm. For g) case, the first layer was considered as having BB origin. QC did not allow the computation of the mean PDR in the layers. For i) case, all three layers have BB origin. However, the QC allowed the estimation of both LR and EAE for first layer and only EAE for the second layer.

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