

## **Response to reviewer #1**

We would like to thank the reviewer for reading this paper attentively and for suggesting both minor corrections and also corrections addressing main science issues. We respond to all his/her comments; the answers are given in blue.

### General comments

**In order to avoid confusions, the same time zone should be used in the figures and throughout the manuscript. Please avoid the use of both LT and UTC. As the instruments used in the study are part of EU and International Infrastructure networks the use of the UTC is preferable.**

Done

**The same unit format should be kept in the manuscript (e.g. m/s or ms<sup>-1</sup>).**

Done

**It is very difficult for the reader to follow the discussion and the Figures when only the case numbering is given. The discussion of the cases as well as the headers of the Figures should be based on the dates of each case or at least the dates and hours of the data should be given together with the case numbering in the plots.**

Dates and hours have been added in Figures 3, 4, 5, 6 and 7. Dates have also been added in the discussion section.

### Introduction

**The authors nicely present the advantages of MAX-DOAS compared to established aerosol measurement techniques (e.g., simple and low cost instrumentation, the ability to perform long-term measurements also in remote areas, the ability to retrieve information on the vertical distribution of aerosol in contrast to sun photometers which only yield AOD) but the shortcomings and the limitations of the technique should also be mentioned in more detail in the introduction.**

We would like to thank the reviewer for this remark. The limitations have been added: i) MAX-DOAS' sensitivity at higher altitudes is low, ii) it provides profiles with much coarser vertical resolution compared to the lidar technique, iii) it performs only daylight measurements

### Section 2.2.2

**One of the major points of the evaluation of the MAX-DOAS aerosol extinction retrievals is the comparison with the extinction lidar profiles. As the study makes use of daytime lidar measurements an assumption of the lidar ratio is needed for the retrieval of the lidar extinction profile. In the manuscript the authors mentioned that the same lidar ratio is used for all cases. Did the authors check the lidar retrievals with e.g. comparison with AOD columnar observation from Cimel?**

We thank the reviewer for this comment. We consider a lidar ratio input value of  $50 \pm 20$  sr (as discussed in Section 2.2.2). This range covers the lidar ratio range for the pollution and dust cases presented in the manuscript (see Fig. 6 in Groß et al. (2013)). This range is also in accordance with columnar lidar ratio values (interpolated to 532 nm) obtained by AERONET for the cases of this study, which vary from  $48.8 \pm 7.5$  sr to  $59.9 \pm 12.1$  sr. The following text is now inserted in the revised manuscript:

“This range is realistic for pollution and dust cases presented herein (Groß et al., 2013) and it is also in accordance with columnar lidar ratio values (interpolated to 532 nm) obtained by AERONET for the cases of this study, which vary from  $48.8 \pm 7.5$  sr to  $59.9 \pm 12.1$  sr).”

Additionally, taking into account the constraints and the assumptions in the lidar ratio, the comparison between lidar-derived AOD and the corresponding AOD values retrieved by MAX-DOAS and AERONET is presented in Figure 7. The lidar AOD errors are now presented in a separate table in the revised manuscript.

AOD (1-4 km)	case (i)- mor	case (i)-aft	case (ii)- mor	case (ii)- aft	case (iii)	case (iv)
lidar	$0.24 \pm 0.04$	$0.21 \pm 0.03$	$0.13 \pm 0.03$	$0.19 \pm 0.03$	$0.19 \pm 0.03$	$0.09 \pm 0.01$
MAX-DOAS	$0.16 \pm 0.03$	$0.15 \pm 0.06$	$0.18 \pm 0.04$	$0.27 \pm 0.05$	$0.19 \pm 0.04$	$0.07 \pm 0.03$

**The uncertainty of the extinction lidar profiles should be discussed, estimated, and given in the manuscript.**

Thank you for raising this point, which was not clear.

Indeed, one of the greatest sources of uncertainty when solving the lidar equation for a common elastic backscatter lidar system is the assumption of a single lidar ratio value, which is considered constant for the entire atmosphere. In the initially submitted manuscript we mentioned that “...the overall uncertainty, including both statistical and systematic errors, on the retrieved  $\beta_{\text{aer}}$  values, is of the order of 20–30% (e.g. Rocadenbosch et al., 2010). In this study, in order to account for the lidar ratio error assumption, we considered a lidar ratio input value of  $50 \pm 20$  sr”. This means that we solved the lidar equation with three different lidar ratio values as input (i.e. 30, 50 and 70 sr) in order to account for this wide variability.

Following the reviewer’s suggestion, the manuscript has been modified accordingly:

“In this study, the aerosol extinction profiles have been retrieved under the assumption of three typical lidar ratio values, 30, 50 and 70 (i.e.  $50 \pm 20$  sr). This range is realistic and in accordance to columnar lidar ratio values (interpolated to 532 nm) obtained by AERONET for the cases of this study, which vary from  $48.8 \pm 7.5$  sr to  $59.9 \pm 12.1$  sr. As a result of this variability (i.e.  $50 \pm 20$  sr), the uncertainties introduced to the aerosol extinction profiles vary from 10 - 40%; the higher uncertainties appear at the upper atmospheric layers, where the signal-to-noise ratio of the system decreases. The corresponding uncertainties for the lidar-derived AOD values of this assumption were estimated to be up to 11%.”

Moreover, the uncertainties of the lidar aerosol profiles are shown in Figure 6 and the uncertainties of the lidar AOD are shown in the table already presented in the previous answer.

**In the L:177-178 the authors stand that the height independent extinction coefficient is representative for the aerosol load in the overlap region, is there any reference that supports this statement? The 1km of the overlap height range is still within the Planetary Boundary layer where an assumption like this could be accepted?**

We thank the reviewer for this comment. Indeed, we agree with the reviewer that the height-independent aerosol extinction coefficient value below 1 km a.s.l. may not be appropriate for the estimation of the aerosol load within the lowest atmosphere, which is mostly affected by the anthropogenic activity. However, this is the best assumption in order to partially account for the aerosol load at the lowest atmospheric layers. The text has been modified accordingly:

“Nevertheless, in order to calculate the AOD from the lidar profiles, the lowermost trustworthy value of the extinction coefficient was assumed constant down to the surface (height-independent). During daytime, the upper limit of the planetary boundary layer over Athens ranges between 1500 and 2100 m a.s.l. (Kokkalis et al. 2020), thus the minimum height of lidar profiles at 1000 m a.s.l. is well within the PBL. Our assumption of a well-mixed atmosphere below 1000 m a.s.l. - which means that a constant lidar ratio value is considered for this part of the atmosphere (Wandinger and Ansmann, 2002) - may lead to an underestimation of the AOD at the lowest troposphere, since the city is most probably a local source of the polluted particles. This underestimation cannot be estimated because of the lidar overlap issue.”

## Section 2.3

**Although already published elsewhere, the general approach and the main features of the algorithm and the optimal estimation method need to be described. For example, a definition of the box air mass factor is missing. What is the a priori aerosol Number concentration profile that is used for the BOREAS retrievals?**

The reviewer is right; we missed to describe important information. The information is now included in the manuscript:

“The algorithm applies the optimal estimation technique for the retrieval of trace gas concentration profiles, while for our case - the aerosol retrievals - it uses an iterative Tikhonov regularization approach. The main concept of the algorithm for the aerosol retrievals is to minimize the difference between modeled and measured O4 slant optical depths by applying the iterative Tikhonov technique to varied aerosol extinction profiles. This method uses the difference of the slant optical depth from an a priori state in order to obtain information on the aerosol concentration that caused this difference through multiple iterations”.

“The BAMF - in contrast to the total AMF - is a function of altitude describing the sensitivity of measurements to the profile at different atmospheric height layers. The aerosol weighting function matrices express the sensitivity of the O4 measurements to changes in the aerosol extinction coefficient profile.”

“...the a priori aerosol number concentration profile  $N_a(z)$  which is used as a starting point for the iterations...”

**The authors should provide further information related to the extinction profiles uncertainties and possible biases in the evaluation with lidar and sun-photometer retrievals due to the a priori selected values.**

We would like to thank the reviewer for the suggestion to address the uncertainties of our calculations in section 2.3. We described the two different errors of our calculations and we calculated the uncertainty introduced to our calculations due to the a priori profile (this information is now included in section 3.3 for each case study separately).

Uncertainties (%)	case (i)-mor	case (i)-aft	case (ii)-mor	case (ii)-aft	case (iii)	case (iv)
smoothing error	15.59	90.52	16.69	13.61	17.46	53.65
noise error	3.94	2.03	2.69	1.93	2.25	5.53

The following text has been added in section 2.3: “The uncertainty associated with each retrieved profile is computed by the algorithm. It is the sum of the noise and smoothing error, which represent the impact of the measurements and of the a

priori profile on the retrieved profile, respectively. These two errors have been calculated for each of our case studies separately and are presented in section 3.2.”

**Table 2 provides information for the input parameters of the 4 selected cases, before the description of the selected cases in the manuscript. Table 2 could provide more generic information, or a rearrangement of the text is needed.**

The reference to Table 2 (renamed to Table 3 in the revised manuscript) has been moved to section 3.1.

### Section 3.1

**A Table providing information (e.g date, atmospheric conditions, air masses), for the 4 selected cases may help the reader to have a better view of the differences and the similarities between the cases. Also, a table will facilitate the reader to follow the discussion which is referring in cases numbering and not in the dates of the cases.**

Thank you very much for this suggestion, which improves the appearance of the manuscript. A new table providing this information has been inserted.

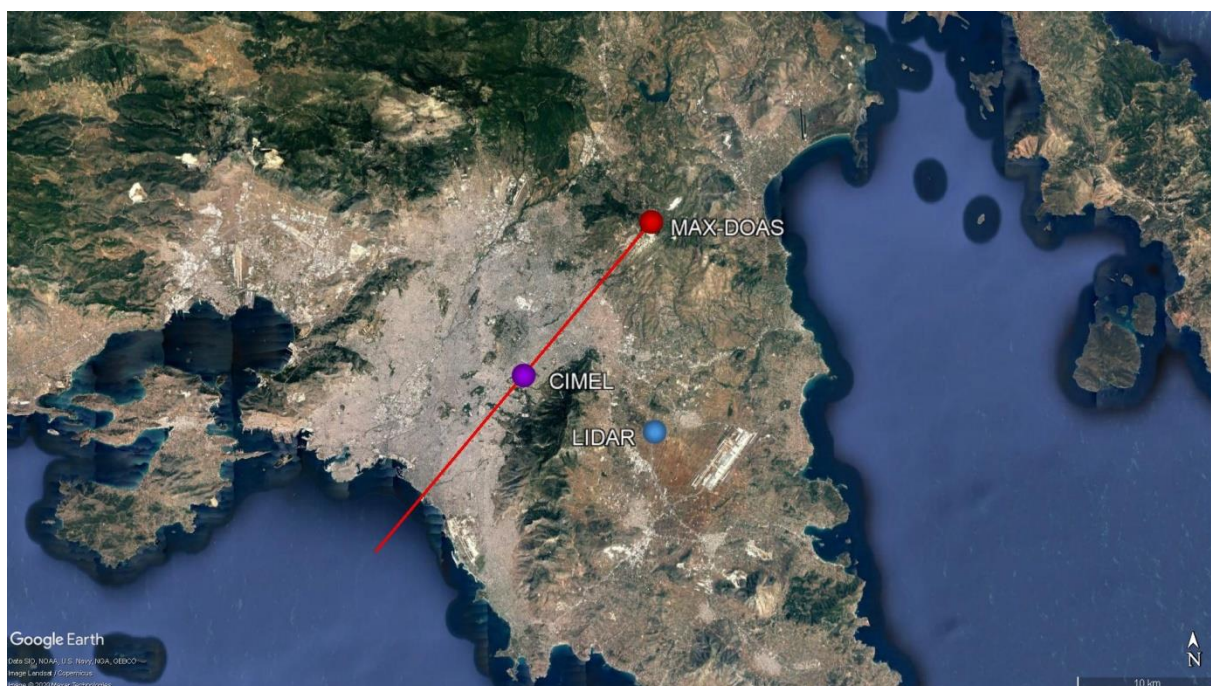
	case (i)	case (ii)	case (iii)	case (iv)
<b>Date</b>	5 Feb 15	9 Jul 15	10 Jul 15	4 Apr 16
<b>Atmospheric conditions</b>	weak dust event, low pollution levels	high pollution levels in the morning	typical pollution levels	high pollution levels
<b>Air masses origin below 4 km</b>	S/SW	N/NE	N/NE	N/NE

**L226: what kind of in situ meteorological observations provide information for cloud free conditions?**

These are empirical observations made by experienced staff of the National Observatory of Athens (NOA) and are registered to the NOA’s official meteorological records.

**Figure 1: A closer map of the area with terrain could better highlight the special topography of the under-study region as well as the orientation and the elevation difference between the instruments. What is the green area in the Figure 1?**

Figure 1 has been updated so that the special topography of the city is more clear now.



## Section 3.2

Since the authors present a technique with main scope to deliver reliable results sensitivity studies are necessary. There is no information how much of the uncertainty of the retrieval is derived from the measurements and how much is from the a priori input.

We would like to thank the reviewer for this comment. The a priori (smoothing) and measurement (noise) errors for each case are now presented in a separate table in the revised manuscript.

Uncertainties (%)	case (i)-mor	case (i)-aft	case (ii)-mor	case (ii)-aft	case (iii)	case (iv)
smoothing error	15.59	90.52	16.69	13.61	17.46	53.65
noise error	3.94	2.03	2.69	1.93	2.25	5.53

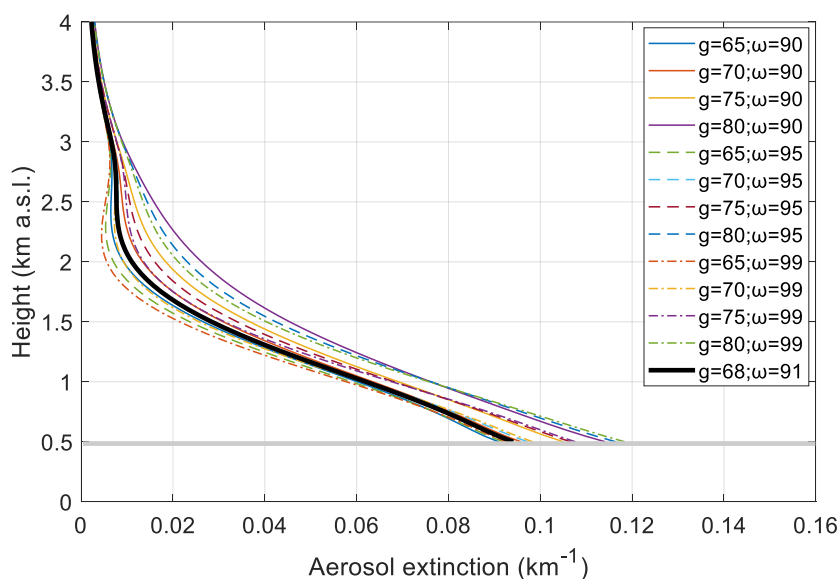
Furthermore, error bars in Figure 6 would help the reader to evaluate the retrieval.

It was an omission from our side, thank you for pointing this out; the errors have been added in the plots of Fig. 6.

Since AERONET measurement are used based on their availability either for the specific date or as a climatological mean value, the resulting uncertainty on the extinction profile should be further discussed and estimated.

Thank you very much for pointing out the influence that the applied aerosol optical properties (SSA and asymmetry factor) have on the algorithm results. For addressing

this, we carried out sensitivity tests with varying SSA and asymmetry factor. The values chosen for the sensitivity test, range between the minimum and maximum values for this season. In the following figure you may see the effect of different SSA and asymmetry factor values on the retrieved profile for case (iv) (which is the only case where a climatological mean was used) at 12:00UTC. The thick black line corresponds to the profile retrieved with the selected parameters for this study ( $g=68$ ,  $\omega=91$ ). The variability due to asymmetry factor is small and the impact of SSA is negligible.



### Section 3.3

**L275: Is the average of more than one extinction profiles, or the average of the lidar signal for the same time window as the MAX-DOAS retrievals? Please be specific.**

The reviewer is right; this point was not clear and is now rephrased: “The lidar profile presented in each figure is the result of the mean lidar signal, averaged between the starting and the ending time of the corresponding MAX-DOAS profiles.”

**L276: Please provide numerical estimation of the uncertainty in the extinction retrievals.**

In L276 of the submitted manuscript (The uncertainty in the lidar extinction profiles increases substantially for altitudes below 1000 m.a.s.l...) we briefly refer to these uncertainties as an explanation for the reason why we use lidar data above that height in our studies. We believe that the numerical estimation of the uncertainty in the extinction retrievals from the lidar measurements below 1000 m a.s.l. is out of the scope of this study. We should emphasize though that this uncertainty does not affect the evaluation of MAX DOAS profiles with the corresponding lidar retrievals, since this is done for heights above 1000 m a.s.l.



**The authors should avoid general and non-specific comments, e.g L292: some discrepancies, L296: some performance statistics. Please rephrase.**

The non-specific comments have been rephrased:

“...hence the retrieved aerosol profiles from the two instruments correspond to different air masses and are not expected to fully agree, especially when the aerosol pollution is not horizontally homogeneous over the Athens basin. Thus, the comparison is mainly focused on a qualitative basis. ”

“Comparison information is given in the form of performance statistics - correlation coefficient ( $r$ ), median lidar/MAX-DOAS ratio, root mean square error (RMSE) and fractional gross error (FGE) – and is shown in Table 4. ”

**L339: an aerosol layer of about 1.5 km deep. In which height?**

It has been rephrased:

“...an aerosol layer extending from the lower atmospheric layers up to 1.5 km height.”

### Section 3.4

#### AOD calculations from Lidar

**In which height range the lidar AOD have been calculated? It is limited to the first 4 km? is there any aerosol layer above 4km that may contribute to the AOD?**

The process followed for the estimation of the AOD from the lidar signal is summarized below:

- (a) For the derivation of range-resolved aerosol optical properties, an aerosol-free reference height window has to be detected initially, where the normalized range-corrected lidar signal fits sufficiently the calculated attenuated molecular backscatter coefficient (Rayleigh-fit criterion; Freudenthaler et al., 2018). For this, the user provides the SCC input platform with an initial guess of that range (in our case 4-6 km based on visual inspection of the range-corrected lidar signal) and the corresponding algorithm fine tunes this guess, by applying different statistical tests to ensure that the shape of the measured signal corresponds to the shape of a Rayleigh signal (Mattis et al., 2016).
- (b) The retrieval of the aerosol backscatter coefficient from the lidar signal starts from the identified reference height using the assumption of the lidar ratio



value. The aerosol extinction coefficient is then calculated from the aerosol backscatter coefficient by multiplying it with the assumed lidar ratio.

- (c) The columnar AOD is derived from the integration of the aerosol extinction profile from ground up to the identified reference height.

No significant aerosol load was observed above the reference height of 4 km a.s.l., in the free troposphere, based on our careful visual inspection of the range-corrected lidar signals. In addition, the AOD variability above the reference height lies within the 3rd decimal place, further ensuring that no significant aerosol layer contributes to the AOD.

Freudenthaler, V., Linné, H., Chaikovski, A., Rabus, D., and Groß, S.: EARLINET lidar quality assurance tools, *Atmos. Meas. Tech. Discuss.*, <https://doi.org/10.5194/amt-2017-395>, in review, 2018

Mattis, I., D'Amico, G., Baars, H., Amodeo, A., Madonna, F., Iarlori, M.: EARLINET Single Calculus Chain – technical – Part 2: Calculation of optical products, *Atmos. Meas. Tech.*, doi:10.5194/amt-9-3009-2016, 9, 3009–3029, 2016

#### AOD evaluation with AERONET

**It is possible the underestimation of the MAX-DOAS to be related to the fact that the AOD from sun photometer is referring to the total column of the atmosphere and the MAX-DOAS covers only the first 4km. Did authors examine the presence of aerosol layers above 4km (e.g lidar observations) for the selected cases? Is this the case for any of the 4 under study cases?**

The reviewer is right, the following brief reference based on his/her comment has been made in the revised manuscript: "...the calculated AOD is limited up to 4 km, while the AOD from CIMEL refers to the total atmospheric column."

The reviewer's suggestion to examine the presence of aerosols at altitudes higher than 4 km could contribute to the observed underestimation by MAX-DOAS; yes, we examined the presence of free tropospheric aerosol layers above the identified reference height (~ 4 km a.s.l.) for all cases with visual inspection of the lidar signal, and no significant aerosol load was observed, as discussed in the previous comment. Although section 3.4 is focused on the comparison with the AOD from CIMEL, the AOD from lidar measurements (calculated by integrating the aerosol extinction coefficient from ground up to the identified reference height of 4 km a.s.l.) is also presented indicatively in the revised manuscript.

**Additionally, the 370m height difference between the location of sunphotometer and MAX-DOAS could have contribution to the AOD differences. This point should further discussed in the manuscript.**

The text has been rephrased accordingly:

“Considering that the sun-photometer is located downtown (150 m a.s.l.), at lower altitude than the MAX-DOAS (527 m a.s.l.) and thus more sensitive to aerosols in the lower troposphere, an underestimation of the contribution of the urban pollution to the retrieved by MAX-DOAS AOD would be expected. Nevertheless, the MAX-DOAS seems to detect well the typical urban aerosols in the boundary layer; the mean AOD difference (CIMEL minus MAX-DOAS) of all the measurements is 0.05 with standard deviation 0.09). ”

The underestimation by MAX-DOAS that is observed in the afternoon is more probably related to MAX-DOAS' viewing geometry as explained in L426-429 in the submitted manuscript.

**Based on the altitude differences and keeping in mind the limitations of lidar to retrieve trustworthy extinction below the full overlap region and the fact that the MAX-DOAS provide profile up to 4km, a comparison of the AOD for the atmospheric layer between 1-4km could provide better conclusions. Is there a reason why this has not been done? The authors should consider to repeat the evaluation of the MAX-DOAS for different altitude ranges.**

We thank the reviewer very much for this remark. He/She is right, the comparison between lidar and MAX-DOAS AOD should be done for a common altitude (1-4 km), since the inclusion of the lowermost atmospheric layer to the AOD calculation adds a lot of uncertainty due to measurement restrictions. The reason for not including this comparison at first was that we would like to focus on the evaluation of the retrieved MAX-DOAS AOD by comparing it with the CIMEL AOD, which is a well established method for AOD retrieval. Nevertheless, we calculated the AOD for 1-4 km altitude and is now presented in a separate table in the revised manuscript.

AOD (1-4 km)	case (i)- mor	case (i)-aft	case (ii)- mor	case (ii)- aft	case (iii)	case (iv)
lidar	0.24 ± 0.04	0.21 ± 0.03	0.13 ± 0.03	0.19 ± 0.03	0.19 ± 0.03	0.09 ± 0.01
MAX-DOAS	0.16 ± 0.03	0.15 ± 0.06	0.18 ± 0.04	0.27 ± 0.05	0.19 ± 0.04	0.07 ± 0.03

#### Section 4

**There is a repetition in 2nd (L:455-463) and 5th (L485-490) paragraph. Please improve the text.**

The reviewer is right; there was a clear repetition, which has been corrected.

**Line466: The authors should be more specific under which atmospheric conditions there is a better agreement. Statements like “in most cases” should be avoid. Please rephrase.**

It has been rephrased:

“...there is good agreement in aerosol layer shape and aerosol extinction levels, except in cases of inhomogeneity at higher altitudes, characteristic of aerosol dust transport episodes. Very good correlation ( $r > 0.84$ ) was found in all cases.”

Technical corrections

**Figure 1: Possible a map with terrain could better highlight the unique topography of the under-study region as well as the orientation and the elevation difference between the instruments. What is the green area in the Figure 1?**

Figure 1 has been updated so that the special topography of the city is more clear now.

**Figure2: Case numbering should be added in the plots.**

Unfortunately this cannot be done since the figures have been generated automatically by the HYSPLIT model. Nevertheless, the dates corresponding to each case have been added in the figure caption.

**Figure 3: The Dates (and hours) should be given as a header in each plot together with the case (i-iv).**

Dates have been added to Figure 3. It has been made more clear in the figure caption that the retrievals shown are the diurnal SC measurements.

**Figure 4: The case (i-iv) should be also given in the plots to facilitate the reader to follow. Please also provide the spatial and temporal analysis of the retrievals in the caption.**

Done.

**Figure 5: The Dates (and hours) should be given as a header in each plot together with the case (i-iv).**

Done.

**Figure 6: The Dates (and hours) should be given as a header in each plot.**

Done.

**Figure 7: The same axis (horizontal and vertical) should be used for each case. Please use the same x-axis (04-17 UTC) for all plots. Maybe a y-axis set at AOD=1.0 will make the plots less busy. Please keep the same format for each plot. The legend of top left plot seems incorrect (e.g lidar 520nm.) Please also mention the date for each case.**

We thank the reviewer for this comment which has improved the appearance of Fig. 7. The figures are now of the same format, with the same y-axes and the date mentioned along with the case number. The reason for keeping the different axes at the internal panels is the better presentation of the scatter plots; when same axes are used, the results are hard to read.

**Table 2: Please correct . . .”Next year’s monthly mean”.**

Rephrased in the caption of Table 2 of the original manuscript: “The mean monthly values of  $\omega$  and  $g$  (provided from AERONET for April 2017) were used for case (iv), due to unavailable AERONET daily data around this date.”