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-1).

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 in lines 67-69 of the revised manuscript: 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 ± 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 ± 7.5 sr to 59.9 ± 12.1 sr. The following text is now inserted in lines 172-174 of 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 ± 7.5 sr to 59.9 ± 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 (Table 7) in the revised manuscript.

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 β_{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 ± 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 following text has been added in lines 171-177 of the revised manuscript:

"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 ± 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 ± 7.5 sr to 59.9 ± 12.1 sr. As a result of this variability (i.e. 50 ± 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 Table 7 of the revised manuscript.

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 in lines 191-196 of the revised manuscript.

"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 and 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 airmass 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 lines 208-213, 217-219 and 226-227 of the revised 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 $N_{\alpha}(z)$ 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 kai 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 Table 5 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

The following text has been added in lines 231-233 in section 2.3 of the revised manuscript: "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 as 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. Table 2 in the revised manuscript is now providing this information.

	case (i)	**case (ii)**	**case (iii)**	**case (iv)**
Date	5 Feb 15	9 Jul 15	10 Jul 15	4 Apr 16
Atmospheric conditions	weak dust event, low pollution levels	high pollution levels in the morning	typical pollution levels	high pollution levels
Air masses origin below 4 km	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 (line 255 of the revised manuscript) 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 in the revised manuscript 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.

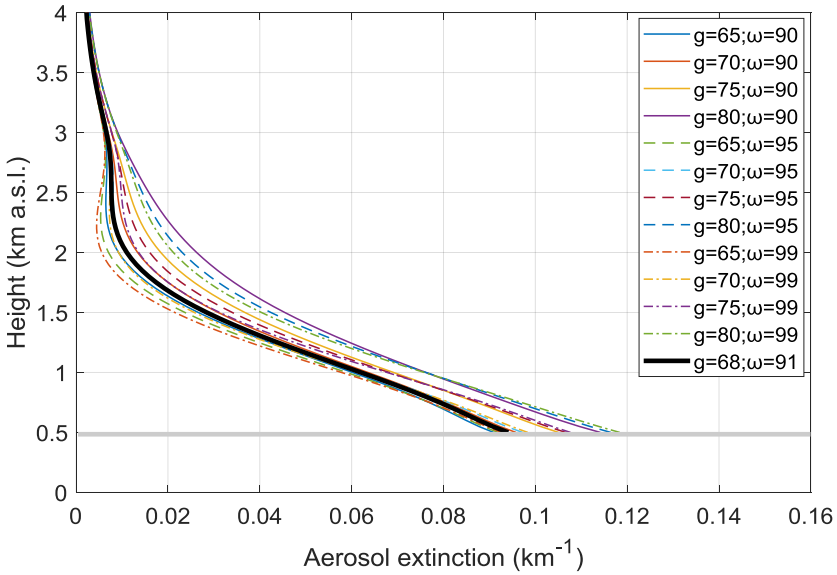
We would like to thank the reviewer for this comment. The a priori (smoothing) and measurement (noise) errors for each case are now presented in Table 5 in the revised manuscript.

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, ω=91). The variability due to asymmetry factor is small and the impact of SSA is negligible. This information is now added in lines 266-267 of the revised manuscript.



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 in lines 311-313 of the revised manuscript: "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 in lines 315-317 and 319-321 of the revised manuscript:

"...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 in lines 364-365 of the revised manuscript:
"...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 in lines 464-469 in the revised version of the manuscript.
"Considering that i) the sun-photometer is located downtown (150 m a.sl.), at lower altitude than the MAX-DOAS (527 m a.s.l.) and thus more sensitive to aerosols in the lower troposphere and ii) the absence of real measurements from MAX-DOAS for altitudes below 500 m a.s.l., 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 0.03 with standard deviation 0.08). "
The underestimation by MAX-DOAS that is observed in the afternoon is more probably related to MAX-DOAS' viewing geometry as explained in lines 426-429 in the originally 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 Table 7 of 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 (lines 503-505 in the revised manuscript):
"...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 ($r > 0.90$) 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.

Figure 2: 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: "The mean monthly values of ω and g (provided from AERONET for April 2017) were used for case (iv), due to unavailable AERONET daily data around this date."

Response to reviewer #2

We would like to thank the reviewer for his/her constructive input and mainly for raising the important issue of the algorithm's performance for the retrievals below station's altitude. We respond to his/her comments; the answers are given in blue.

The study "Retrieval and evaluation of tropospheric aerosol extinction profiles using MAX-DOAS measurements over Athens, Greece" presents aerosol profiles resulting from an inversion of MAX-DOAS measurements with the BOREAS algorithm. It generally matches the scope of AMT. However, there are major shortcomings in the methodology which require major clarifications, additional RTM calculations, and extended discussions. The manuscript should thus not be accepted for AMT unless major extensions and revisions are made. Thus, the current review only focusses on the retrieval part, as the presented results are likely going to change.

Retrieval shortcomings

1. Observation geometry

MAX-DOAS profile retrievals have been developed and refined in the last years and have been shown to yield valuable information on trace gas and aerosol profiles. However, the assumptions made in BOREAS (as well as other inversion schemes) put a MAX-DOAS instrument at the ground within flat terrain.
The situation in Athens seems to be quite different: the instrument is located at a hillside at _500m altitude. SCDs at negative and zero elevation angles have been measured, but are not included in the analysis. So I wonder how the near-surface extinction in Athens could be derived from an instrument on a hill looking upwards!? This aspect is not really explained and discussed quantitatively in the current manuscript.

We would like to thank the reviewer for his critical remarks which helped us to realise a misinterpretation of our results. Indeed, the station's location is unusual and the retrieval of extinction values below station altitude needs further discussion. In principle, some light reflected on thesurface and scattered in the atmosphere below the station altitude will be scattered also into upward pointing line of sights in particular for the lowest elevation angles. However, over dark surfaces this is a relatively small contribution to the total intensity and thus carries limited information on extinction in the lowest layers. In addition, the RTM SCIATRAN is a 1d model and cannot account for any effects related to the complex topography.

In response to the reviewer's comments, we have performed a series of sensitivity studies performing retrievals on synthetic data created using different vertical profiles of aerosol extinction. The results indicate that the extinction retrieved below station altitude is dominated by the a priori, scaled to the total retrieved AOD. In view of these disappointing results, we have revised the manuscript by removing all results below station altitude and changing the discussion accordingly.

I am not aware of a MAX-DOAS publication with similar viewing geometry. As the current study seems to be the first, it could be pioneering in this aspect.

MAX-DOAS Measurements from elevated stations have been reported in a number of publications, including Gomez et al., 2014, Schreier et al., 2016, Bognar et al., 2020, Ma et al., 2020, Wang et al., 2020). However, with the exception of Bognar et al. and Wang et al., they do not attempt a full profile retrieval.

Gomez, L., Navarro-Comas, M., Puentedura, O., Gonzalez, Y., Cuevas, E., and Gil-Ojeda, M.: Long-path averaged mixing ratios of O3 and NO2 in the free troposphere from mountain MAX- DOAS, Atmos. Meas. Tech., 7, 3373–3386, doi:10.5194/amt-7- 3373-2014, 2014

Schreier, S. F., Richter, A., Wittrock, F. and Burrows, J. P.: Estimates of free-tropospheric NO2 and HCHO mixing ratios derived from high-altitude mountain MAX-DOAS observations at midlatitudes and in the tropics, Atmos. Chem. Phys., 16(5), 2803–2817, doi:10.5194/acp-16-2803-2016, 2016.

Bognar, K., Zhao, X., Strong, K.,Chang, R. Y.-W., Frieß, U.,Hayes, P. L., et al. (2020).Measurements of tropospherichromine monoxide over four halogenactivation seasons in the Canadianhigh Arctic. Journal of GeophysicalResearch: Atmospheres, 125,e2020JD033015. https://doi.org/10.

Ma, J., Dörner, S., Donner, S., Jin, J., Cheng, S., Guo, J., Zhang, Z., Wang, J., Liu, P., Zhang, G., Pukite, J., Lampel, J., and Wagner, T.: MAX-DOAS measurements of NO2, SO2, HCHO, and BrO at the Mt. Waliguan WMO GAW global baseline station in the Tibetan Plateau, Atmos. Chem. Phys., 20, 6973–6990, https://doi.org/10.5194/acp-20-6973-2020, 2020.

Wang, Z., Chan, K. L., Heue, K.-P., Doicu, A., Wagner, T., Holla, R., and Wiegner, M.: A multi-axis differential optical absorption spectroscopy aerosol profile retrieval algorithm for high-altitude measurements: application to measurements at Schneefernerhaus (UFS), Germany, Atmos. Meas. Tech., 13, 1835–1866, https://doi.org/10.5194/amt-13-1835-2020, 2020.

But it has to provide far more details, discussion and RTM calculations in order to interprete the resulting BOREAS profiles:
- what is the exact setup for the BOREAS retrieval? I assume the instrument was set to station altitude. But where is the surface in RTM calculations? At station level aswell? At sea level? I assume that simulating 3D terrain is not easily possible, but the potential impact of terrain should at least be discussed.

Indeed, the instrument was set to station's altitude in the RTM calculations. The surface was set at sea level (this is now mentioned in lines 222-223 of the revised manuscript). As mentioned above, SCIATRAN is a 1d model, which does not allow modeling the effects of a variable topography.

- how large is the MAX-DOAS sensitivity for aerosols below station height? Fig. 5 indicates high sensitivity to altitudes below station level, but I really wonder where this should be coming from as the instrument is only looking upwards.

As discussed above, we have tested the sensitivity using synthetic data. As it turned out, the sensitivity is very low and the results are thus dominated by the a priori profile shape. This was not correctly reflected in the averaging kernels shown in Fig. 5 as for the atmospheric layers below station's height, the value for 500 m was shown. This has been corrected in the revised manuscript.

- what is the meaning of the aerosol contour plots down to sea level, and where is the information coming from?

As this information was mainly coming from the a priori, we have decided to remove it from Fig. 4 in the revised manuscript, which now presents only retrievals from instrument's height up to 4 km a.s.l.

- how has the AOD derived by BOAS to be interpreted? Is it really the full AOD for the city of Athens (where the MAX-DOAS is pointing at), or just the fractional AOD from station altitude upwards?

In the originally submitted manuscript, the AOD from BOREAS was the integration of the extinction values from the surface up to 4 km. In the revised paper, we present both the fractional AOD (1-4 km) in Table 7 of the revised manuscript and the AOD for the whole column by setting a constant value (equal to the retrieved value at 500 m) for the lower levels, where the retrievals are not trustworthy (Fig. 7).

- from the contour plot, I would conclude that the largest fraction of the extinction profile is below 500 m. This could also be expected for urban pollution accumulating in a valley. But in order to interprete these results and the contour plots, it is essential to give evidence (by RTM calculations) that aerosol profiles can be actually derived down to the ground from an elevated intrument looking upwards. If the extinction below station altitude cannot be trusted, however, it should be discarded from Fig. 4. In this case, the comparisons should also only consider the fractional AOD from station altitude upwards. The case studies would than have to be revised completely.

We agree with the reviewer that the MAX-DOAS retrievals of the extinction coefficient for heights below 500 m need to be optimized and that the measurements with negative elevation angles need to be included in the algorithm retrievals. However, for technical reasons, the measurements at negative elevation angles can currently not be included in the retrieval, even if good knowledge of thesurface albedo is assumed. As discussed above, we performed several RTM simulations with synthetic data in order to assess the sensitivity for aerosol layers below 500m (for which in our case using only upward observations, the information comes only from multiple scattering). However, the results were discouraging and we therefore decided to follow the reviewer's suggestion to exclude the retrievals below 500 m in the revised manuscript. The results excluding the retrievals of the lower layers are shown in the revised Figure 4. We are not attaching the results from the RTM calculations in this reply, but they are at your disposal if needed.

Regarding the AOD calculation, the missing values in the extinction coefficient profiles below 500 m are set to a constant value (equal to the retrieved value at 500 m). This assumes that the atmosphere is well-mixed below 500 m, which probably results in an underestimation of the calculated AOD in case of enhanced surface aerosol layer. In the revised manuscript we provide the new AOD calculations for

MAX-DOAS in Figure 7, along with their comparison with the sun-photometer AOD in Table 6.

2. Standard atmosphere

The profiles used for T and p affect the O4 VCD, which affects the aerosol profile inversion. Using a standard atmosphere is thus not appropriate. I recommend to repeat the analysis with more realistic T/p profiles for Athens. At least, the authors have to quantify the effect of using a standard atmosphere for winter vs. summer.

We would like to thank the reviewer for this important suggestion. We rerun all the retrievals using measured T/p profiles for Athens from the Atmospheric Science Radiosonde Archive of the University of Wyoming (http://weather.uwyo.edu/upperair/bufrraob.shtml) (lines 220-222). We provide all the profiles and statistics anew in the revised manuscript.

Minor comments:

Eq. 1: It might be worth mentioning that SCD_alpha is actually rather SCD_alpha-SCD_90 as well, as the zenith SCD has been used as reference in the DOAS analysis.

Thank you very much for pointing out this omission; we now made it more clear in the revised manuscript:

$$VCD = \frac{SCD_{\alpha} - SCD_{90^o}}{(AMF_{\alpha} - AMF_{90^o})}$$

Page 8, line 234 and line 237: This information should already be given in section 2.2.1.

Thank you for this remark, we agree and this information has been moved to section 2.2.1 (lines 116-118).

Marked-up manuscript version

Retrieval and evaluation of tropospheric aerosol extinction profiles using MAX-DOAS measurements over Athens, Greece

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Abstract. In this study, we report on the retrieval of aerosol extinction profiles from ground-based scattered sunlight multi-axis differential optical absorption spectroscopy (MAX-DOAS) measurements, carried out at Athens, Greece. It is the first time that aerosol profiles are retrieved from MAX-DOAS measurements in Athens. The reported aerosol vertical distributions at 477 nm are derived from the oxygen dimer (O_4) differential slant column density observations at different elevation angles by applying the BOREAS retrieval algorithm. Four case studies have been selected for validation purposes; the retrieved aerosol profiles and the corresponding aerosol optical depths (AODs) from the MAX-DOAS are compared with lidar extinction profiles and with sun photometric measurements (AERONET observations), respectively. Despite the different approach of each method regarding the retrieval of the aerosol information, the comparison with the lidar measurements at 532 nm reveals a very good agreement in terms of vertical distribution, with r>0.90~~r>0.85~~ in all cases. The AODs from the MAX-DOAS and the sun-photometer (the latter at 500 nm) show a satisfactory correlation (with 0.45 < r < 0.7~~r≈0.6~~ in three out of the four cases). The comparison indicates that the MAX-DOAS systematically underestimates the AOD in the cases of large particles (small Ångström exponent) and for measurements at small relative azimuthal angles between the viewing direction and the Sun. Better agreement is achieved in the morning, at large relative azimuthal angles. Overall, the aerosol

profiles retrieved from MAX-DOAS measurements are of good quality; thus, new perspectives are opened up for assessing urban aerosol pollution on a long term-basis in Athens from continuous and uninterrupted MAX-DOAS measurements.

1 Introduction

Tropospheric aerosols originate from both natural and anthropogenic sources. The lifetime of aerosols in the troposphere ranges from a few days to a few weeks, depending on their size and meteorology (e.g. Pandis et al., 1995). They take part in atmospheric processes through (i) nucleation and interaction with clouds (e.g. Twomey et al., 1977; Rosenfeld et al, 2014), (ii) participation in chemical and photochemical reactions, by providing the required surface for heterogeneous reactions to take place (Andreae & Crutzen, 1997) and (iii) absorption and scattering of incoming solar and earth's IR radiation, affecting atmospheric dynamics and stability (e.g. Dubovik et al., 2002) and the Earth's climate (IPCC, 2001). Significant decrease of UV-Vis irradiance reaching the ground due to urban aerosol pollution has been reported in various cases (e.g. Zerefos et al., 2009; Chubarova et al., 2011).

According to a survey conducted in 25 large European cities, Athens occupies the third position on European level in exceedances of particle pollution regulations (Pascal et al., 2013). Saharan dust transported from the African continent is the main natural source of tropospheric aerosols in Athens (e.g. Kanakidou et al., 2007; Gerasopoulos et al., 2011; Raptis et al., 2020), while common anthropogenic sources are traffic emission and domestic heating (Markakis et al, 2010; Gratsea et al., 2017). Wildfires also contribute to the aerosol mixture in the area occasionally, either from local~~by nearby~~ events (Amiridis et al., 2012) or by long-range transport (Papayannis et al.; 2009, Amiridis et al., 2011; Mona et al., 2012). Whereas emissions of most air pollutants, such as SO_2, are expected to decrease by more than 80% until the end of the 21st century, the decrease of aerosol emissions is projected to be small (IPCC 2007) and thus aerosols may play an even more critical role in air quality in the future. Therefore, long-term continuous measurements, providing information on the spatial and temporal distribution of aerosols, are of great importance to urban air pollution assessment and to the understanding of the aerosol contribution to Earth's climate. The knowledge of the vertical distribution of aerosols is necessary for understanding the mechanisms underlying the formation and development of urban smog.

Satellite, airborne and ground-based measurements are widely used to derive aerosol vertical profiles (e.g., Papayannis et al., 2005; Schmid et al., 2006; DeCarlo et al., 2008; Solanki and Singh, 2014); satellite measurements sometimes fail to be accurate in the lower atmosphere, while airborne measurements, although accurate in the lower atmosphere, are temporally restricted. In contrast, ground-based measurements can provide both a very good record of the lower troposphere and a satisfactory temporal resolution. However, since the ground-based profile measurements are mainly relying on lidar systems (e.g., the European Aerosol Research Lidar Network - EARLINET – within the European Research

Infrastructure for the observation of Aerosol, Clouds and Trace Gases - ACTRIS), they are costly in terms of setup and operation. An additional option for ground-based observations is the MAX-DOAS technique, which has been gaining ground over the last years (e.g., Wittrock et al., 2004; Heckel et al., 2005, Ma et al., 2013, Schreier et al., 2020) since it can provide low-cost, continuous and uninterrupted measurements without the need for absolute radiometric calibration. The MAX-DOAS technique has also been shown to be very promising for the retrieval of aerosols' vertical distribution (e.g., Sinreich et al., 2005; Lee et al., 2009; Clémer et al., 2010; Wagner et al., 2011). However, its sensitivity at higher altitudes is low and compared to the lidar technique, it provides profiles with much coarser vertical resolution. It also performs only daylight measurements, which can be considered as a limitation of this technique. In some studies, the retrieved aerosol extinction profiles from MAX-DOAS measurements are~~were~~ compared to the corresponding profiles derived from lidar (e.g., Irie et al., 2008; Zieger et al., 2011; Bösch et al., 2018) or Aerosol Robotic Network (AERONET) based measurements (e.g. Wang et al., 2016). For the Athens area, although several studies have been published on aerosol extinction profiles from lidar measurements (e.g. Papayannis et al., 1998; Matthias et al., 2004; Papayannis et al., 2005), vertical trace gas and aerosol profile retrievals from MAX-DOAS have not been published so far.

In the scope of this paper, a retrieval algorithm, recently developed by the Institute of Environmental Physics and Remote Sensing of University of Bremen (Bösch et al., 2018), is employed in order to obtain vertical distributions of aerosol extinction from O_4 MAX-DOAS measurements over the urban environment of Athens. O_4 is an atmospheric absorber with a known concentration profile, therefore ~~its absorption~~ measurements of the O_4 column can be used to retrieve~~as an indicator of~~ the aerosol induced light path changes (Wagner et al., 2004).

For validation purposes the outcomes of our calculations are compared to established techniques; the retrieved profiles are compared to profiles from ground-based lidar measurements (EARLINET station) and the AOD to sun-photometer measurements (AERONET station).

A description of the instruments used in this study (location, instrumentation and data retrieval) along with a brief description of the profile retrieval algorithm are given in section 2. In section 3, we present the derived aerosol vertical distributions for four selected case studies and we compare the MAX-DOAS aerosol extinction coefficient profiles and the AOD with lidar and sun-photometric measurements, respectively. The findings are summarised in section 4, where also the conclusions of this study are provided.

2 Methodology

2.1 Location

Four mountains surround the city of Athens, forming a basin that is open to the south and southwest. This special topography plays an essential role in the accumulation of atmospheric pollutants over the city under certain meteorological conditions (Kassomenos et al., 1995). Moreover, dust transport episodes from North Africa also contribute to the aerosol load of the city (e.g. Gerasopoulos et al., 2009; Kosmopoulos et al., 2017). In general, the Athens area can be considered as an example of various aerosol types such as dust, local pollution, marine, biomass combustion and their mixtures (Soupiona et al., 2019).

Figure 1 shows the Greater Athens area and the location of each instrument used in this study. The MAX-DOAS instrument is located at the premises of the National Observatory of Athens (NOA, 38.05° N, 23.86° E, 527m a.s.l), to the north of the city. No strong emission sources are present around the measurement area, which is considered as suburban background. The lidar system performs measurements at the National and Technical University of Athens (NTUA, 37.97 °N, 23.79 °E, 212m a.s.l.) and the site is considered as suburban background. The CIMEL sun-photometer is installed at the premises of NOA at Thissio hill (37° 58′ N, 23° 43′ E, 150m a.s.l.), which, despite being located in the city centre, is considered as urban background (Paraskevopoulou et al., 2015). Information about the instruments is provided in Table 1.

2.2 Instrumentation and data retrieval

2.2.1 MAX-DOAS

The MAX-DOAS instrument employed in this study is part of the BREDOM network (Bremian DOAS network for atmospheric measurements, http://www.iup.uni-bremen.de/doas/groundbased_data.htm) and has been operating continuously since October 2012. It comprises a grating spectrometer (LOT 260S, 600 l/mm ruled grating) connected via an optical fiber bundle to a computer-controlled telescope unit. The spectrometer covers a spectral range from 330 to 500 nm with a spectral resolution of approximately 0.7 nm. The detector used is a CCD (Charge-Coupled Device) by Andor Technology, with 2048 x 512 pixel resolution, cooled to -40°C.

The telescope performs intensity measurements at eight elevation angles (-1°, 0°, 1°, 2°, 4°, 8°, 15°, 30°), as well as to the zenith. However, the current retrieval algorithm only considers upward viewing directions, excluding the -1° and 0°. With this choice, little information is available for the profile retrieval below the station altitude, therefore profiles are retrieved and presented only for altitudes above 500 m a.s.l. Measurements in eight azimuthal directions are performed, but in this study, only the S direction -~~,~~ pointing at 52.5° (with respect to South) and associated to~~representing~~ the urban atmospheric conditions~~area~~ of the city (Gratsea et al., 2016) -~~,~~ is considered (Fig. 1). The S direction also covers the sun-

photometer's location and points close to the lidar's measurement site. The duration of one full scanning cycle (azimuthal and elevation scanning) is about 15 min, thus about 30 measurement cycles per day are available in winter and 45 in summer.

The spectral measurements are analysed using the DOAS technique; the Beer-Lambert law is considered as the solution of the radiative transfer equation (Platt and Stutz, 2008)~~(Hönninger and Platt, 2002; Wittrock et al., 2004)~~ and the absorption spectrum is separated into broad and narrow spectral features that show low and high frequency variations, respectively, as a function of wavelength. The narrow spectral features correspond to the unique narrow-band absorption structures of the trace gases, while the broad ones represent the attenuation of solar radiation by scattering processes in the atmosphere as well as the continuum absorption by trace gases and the instrument. For the derivation of the slant column density (SCD, defined as the concentration of the absorber integrated along the light path), a polynomial accounting for the broad spectral features and the laboratory cross-sections of the retrieved species are fitted to the measured optical depth. To determine the optical depth, the logarithm of the ratio of the current horizon measurement (I) and the reference intensity (I_o) is taken.

The SCD of the oxygen dimer (O_4), i.e. the slant optical thickness of the absorber divided by the absorption cross section, measured at different elevations is used as input to the retrieval algorithm for the calculation of the aerosol distribution. The slant column of the O_4, a weak molecular absorber with a well-known vertical profile (the O_4 concentration is proportional to the square of the O_2), is almost linearly dependent on the average photon pathlengths (Pfeilsticker et al., 1997) and thus can be used as an indicator of the presence of clouds or aerosols in the atmosphere. The SCD_{O4} is calculated by fitting to the measured optical depth the laboratory spectrum of O_4 (Hermans et al., 2003), NO_2 (Vandaele et al., 1998) and of O_3 (Bogumil et al., 2000) and a polynomial of degree 4 which accounts for the broad spectral features. The fitting spectral window used is 425-490 nm. In order to retrieve the tropospheric SC_{O4}, the zenith observation, corresponding to each measurement cycle, is used as the reference measurement I_o, canceling in this way the Fraunhofer lines in the solar spectrum and the stratospheric contributions to the SCD.

Tropospheric~~The differential tropospheric~~ vertical column densities (VCD) of NO_2, shown in section 3.1, can be derived by using air mass factors (AMF) calculated with the SCIATRAN radiative transfer model (Rozanov et al., 2000). To convert the differential tropospheric SCD to the corresponding tropospheric VCD, the differential AMF ($AMF_\alpha - AMF_{90^o}$) is required, namely the difference between the AMF at the same elevation α as the SCD measurement and the AMF at the zenith (Eq. 1).

$$VCD = \frac{SCD_\alpha - SCD_{90^o}}{(AMF_\alpha - AMF_{90^o})} \quad \sout{VCD = \frac{SCD_\alpha}{(AMF_\alpha - AMF_{90^o})}}$$

(1)

The AMF describes the weighting of the absorption as a function of the relative azimuth and the solar zenith angle (SZA) for a given atmospheric profile and at a specific wavelength.

2.2.2 EOLE lidar system

The six-wavelength Raman-backscatter lidar system (EOLE) operates in Athens since February 2000 as part of the EARLINET network (Pappalardo et al., 2014). The system is designed following the optical set-up of a typical member station (Kokkalis 2017), meeting all the quality assurance requirements of the network. The emission unit is based on a pulsed Nd:YAG laser, emitting high energy pulses at 355, 532 and 1064 nm with a repetition rate of 10 Hz. The optical receiver is based on a Cassegrainian telescope (600 mm focal length and a clear aperture diameter of 300 mm), directly coupled with an optical fiber, to the wavelength separation unit, detecting signals at 355, 387 (N_2 Raman line of 355nm), 407 (H_2O Raman line of 355nm), 532, 607 (N_2 Raman line of the 532nm) and 1064 nm. For every measuring cycle 1000 lidar signal returns are stored (every~1.66). For each case presented in this study, we used hourly averaged profiles, which correspond to approximately 34 individual signal acquisitions (Kokkalis et al., 2012).

During day time operation, the system is capable of providing aerosol backscatter profiles (β_{aer}) at 355, 532 and 1064 nm, based on the standard backscatter lidar technique and employing the Klett inversion method (Klett, 1981). This technique assumes the existence of an aerosol-free region (e.g. upper troposphere) and requires an a-priori assumption of the lidar ratio value (the ratio of the extinction to backscatter coefficient, S_{aer}). A variety of studies revealed a wide range for the lidar ratios, covering values from 20 to 100 sr (Ackermann, 1998; Mattis et al., 2004; Amiridis et al., 2005; Müller et al., 2007; Papayannis et al., 2008; Groß et al., 2011; Giannakaki et al., 2015). When the elastic backscatter lidar technique is used, the assumption of a constant lidar ratio value throughout the laser sounding range, becomes very critical when solving the lidar equation; in this case, the overall uncertainty, including both statistical and systematic errors, on the retrieved β_{aer}values, is of the order of 20–30% (e.g. Rocadenbosch et al., 2010). In this study, the aerosol extinction profiles have been retrieved under the assumption of three lidar ratio values, 30, 50 and 70 (i.e. 50 ± 20 sr). 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 ± 7.5 sr to 59.9 ± 12.1 sr. As a result of this variability (i.e. 50 ± 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 of the lidar-derived AOD values due to this assumption were estimated to be up to 11%. All the lidar profiles were~~In this study, in order to account for the lidar ratio error assumption, we considered a lidar ratio input value of 50 ± 20 sr. All the lidar profiles where~~ obtained with the Single Calculus Chain (SCC) processing platform (D'Amico et al., 2016; Mattis et al., 2016), which is developed in the framework of EARLINET to ensure the high-quality products of the network, by implementing quality checks on both raw lidar data and final optical products.

One of the lidar's main limitations is the distance of full overlap between the laser beam and the receiver's field of view, which makes it difficult for the instrument to obtain useful and accurate aerosol-related information below that height. Wandinger and Ansmann (2002) demonstrated that when not applying overlap correction in lidar signals, the retrieved aerosol extinction coefficient may take even non-physical negative values for heights up to the full overlap. The incomplete overlap effect can be solved by using Raman measurements under night-time conditions. In this study, only daytime measurements are used and therefore no overlap correction is applied on the signals. The geometrical configuration of EOLE results in full overlap distance of 500-800 m above ground (Kokkalis 2017). The aerosol extinction values below the 1000 m a.s.l. height are considered to be inside the overlap region and therefore were omitted from the extinction profile comparison. Nevertheless, in order to calculate the AOD from the lidar profiles, the lowermost trustworthy value of the extinction coefficient was assumed constant (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 an additional source of particles. This underestimation cannot be estimated because of the lidar overlap issue.~~(height-independent), and also representative of the aerosol load in the overlap region.~~

2.2.3 CIMEL sun-photometer

The reported columnar aerosol optical properties have been retrieved by a CIMEL sun-photometer (Holben et al., 1998). The instrument is part of NASA's global sun photometric network, AERONET, and performs automatic measurements of the direct solar radiance at the common wavelengths of 340, 380, 440, 500, 675, 870, 940 and 1020 nm every 15 min and diffuse sky radiance at 440, 675, 870 and 1020 nm. These measurements are further used to provide both optical and microphysical aerosol properties in the atmospheric column (Dubovik et al., 2006). The CIMEL data used in this study are the cloud screened and quality assured level 2.0 data products, providing information about the columnar AOD and the Ångström exponent. The AOD uncertainty is <±0.02 for UV wavelengths and < ±0.01 for wavelengths larger than 440 nm (Eck et al., 1999).

2.3 BOREAS profile retrieval algorithm

The BRemen Optimal estimation REtrieval for Aerosol and trace gaseS (BOREAS) is an optimal estimation based profile retrieval algorithm developed at the Institute of Environmental Physics, University of Bremen (Bösch et al., 2018). It 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 retrieval is to minimize the difference between modeled and measured O_4 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 to obtain information on the aerosol concentration that caused this difference through multiple iterations. Slant column densities of trace gases~~Slant column densities of NO_2~~ and O_4 from MAX-DOAS measurements at different line of sight (LOS) directions, as well as climatology profile files are used as inputs. The BOREAS algorithm is based on the SCIATRAN radiative transfer model (Rozanov et al., 2005), which is used to calculate box-air-mass-factors (BAMF) and weighting functions, needed for the profile inversion. 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 O_4 measurements to changes in the aerosol extinction coefficient profile. For the radiative transfer model (RTM) calculations, scattered light in a spherical atmosphere (multiple scattering) and atmospheric profiles of pressure and temperature for Athens from the Atmospheric Science Radiosonde Archive of the University of Wyoming (http://weather.uwyo.edu/upperair/bufrraob.shtml) are considered. The instrument was set to station's altitude and the surface was set at sea level.~~from the U.S. Standard Atmosphere (NASA, 1976) are considered.~~ The aerosol inversion problem is expressed through the minimisation of Eq. (2):

$$\left\| \Delta\tau(\lambda, \boldsymbol{\Omega}) - \Delta\tilde{\tau}\left(\lambda, \boldsymbol{\Omega}, N_\alpha(z)\right) - P(\lambda, \boldsymbol{\Omega}) \right\|^2 \rightarrow min \quad (2)$$

,where Δτ denotes the measured O_4 differential slant optical thickness, $\Delta\tilde{\tau}$ the simulated differential slant optical thickness, $\boldsymbol{\Omega}$ the measurement geometry (LOS, SZA, relative azimuth) , $N_\alpha(z)$~~(z)~~ the a priori aerosol number concentration profile which is used as a starting point for the iterations and P a polynomial of lower order which accounts for the attenuation due to scattering processes.~~.~~ Since the relationship between the concentration profile and the O_4 differential slant optical depth is not linear, the~~an~~ iterative Tikhonov regularisation technique, along with weighting function matrices, is ~~expressing the sensitivity of the measurements to changes in the aerosol profile, are~~ used for the solution of the minimisation~~optimisation~~ problem (Bösch et al., 2018).~~.~~

The uncertainty associated with each retrieved profile is computed by the algorithm. It is the sum of the noise and smoothing errors, 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.

The temporal resolution of the measurements is about 15 minutes, which corresponds to the duration of one full scanning cycle through all directions over the city. The vertical sampling of the retrieved profile is 0.05 km, with the bottom layer considered at the sea level and the top layer at 4 km a.s.l. The AOD is calculated by integrating the BOREAS retrieved aerosol extinction coefficient vertically. ~~Table 2 summarises the parameter settings used for the BOREAS retrieval.~~ More details about the values assigned to each parameter are given in section 3.2.

3 Results and discussion

3.1 Selected case studies

The main objective of this study is to assess the retrieved aerosol profiles from MAX-DOAS measurements by comparing them with well established sun-photometric measurements (CIMEL) and lidar retrievals. Therefore, certain cases had to be selected with available and valid data from all three instruments. Additionally, the selected cases had to coincide with cloud-free days, as all of the used measurement techniques have more substantial~~subtantial~~ uncertainties in the presence of clouds. During the period from January 2015 to June 2016, four cases were found to meet the above conditions, covering winter, summer and spring: i) 05 February 2015 under the influence of a weak dust event, ii) 09 July 2015 with enhanced morning levels of NO_2 for this season (Gratsea et al., 2016) iii) 10 July 2015 with typical levels of pollution and iv) 04 April 2016 with enhanced levels of NO_2. In order to identify the sources of air masses reaching Athens on the specific dates, 4-day air mass back trajectories at different altitudes, calculated using the NOAA-HYSPLIT (Hybrid Single-Particle Lagrangian-Integrated Trajectory) model (Draxler and Hess, 1997) were used. Potential for Saharan Dust transport below 4 km, which is the highest point of our retrievals, was identified only for case (i) (Fig.2). In the rest of the cases, the air masses below 4 km originate from N/NE directions, and are thus not associated with dust aerosols. The NO_2 levels, measured by MAX-DOAS and presented in Fig. 3, are used as an indicator for the pollution levels over the city. The mean diurnal NO_2 DSCDs for winter and summer months, as reported by Gratsea et al. (2016), range from $6 \cdot 10^{16}$ to $9 \cdot 10^{16}$ and $5 \cdot 10^{16}$ to $11 \cdot 10^{16}$ molec·cm^{-2},~~molec/cm^2~~, respectively. Thus, enhanced pollution levels are observed during the morning hours in cases (ii) and (iv). The absence of clouds is established using in-situ empirical meteorological observations from the monitoring station of the National Observatory of Athens at the centre of the city and is also verified by the MAX-DOAS retrieved O_4 slant columns throughout the day. The above mentioned cases will henceforth be referred to as cases (i), (ii), (iii) and (iv), respectively and information about each case is summarised in Table 2.~~.~~

~~The MAX-DOAS elevation angle sequence includes not only measurements above the horizon, but also at 0° and -1° elevation. Initially, the aerosol extinction profiles from MAX-DOAS were derived using measurements including these elevation angles. However, experience showed that better results are achieved only when elevation angles above the horizon are used for the inversion, most probably due to the negative impact of an inaccurate spectral surface reflectance on the retrieval. Thus the -1° and 0° elevation angles were excluded from the calculations. With this choice, little information is available for the profile retrieval below the station altitude of 527 m a.s.l. As already mentioned (Fig. 1), the S azimuthal direction is associated to the city's urban atmospheric conditions (Gratsea et al., 2016). The S direction also coincides with the sun-photometer's location and is also close to the lidar's measurement site (Fig.1).~~

3.2 Aerosol extinction vertical profile retrievals

MAX-DOAS measurements and the BOREAS retrieval algorithm were used for the calculation of the diurnal aerosol extinction vertical distribution over the urban (S) area (Fig. 4) for the selected case studies and for altitudes 0.5-4 km a.s.l.~~.~~ Single scattering albedo (SSA) and phase functions are not retrieved in BOREAS and have to be prescribed. Therefore AERONET measurements are used for specifying SSA (ω) and asymmetry factor (g) values. However, ω and g were not available in AERONET data for case (iv), therefore in this case the algorithm was run using the Henyey-Greenstein phase function with the monthly mean of SSA (ω=0.91) and asymmetry factor (g=0.68) from the following year, as derived from the AERONET data (Table 3). Specifically for this case, sensitivity tests with varying ω and g were carried out. It was found that the variability due to asymmetry factor is small and the impact of SSA negligible.~~2).~~ A fixed surface albedo (α=0.15), based on a previous study for Athens (Psiloglou et al., 2009), was used in all cases. Table 3 summarises the parameter settings used for the BOREAS retrieval.

The results for case (i) reveal a significant variation of the aerosol distribution in the vertical direction. Although the ~~The~~ maximum retrieved extinction values in this case~~,~~ reach almost 0.2~~0.3~~ km^{-1} at 1.5 km height in the afternoon, persistent high values are displayed~~close to the surface~~ until around local noon (Fig. 4). The~~and after that time the aerosol load declines. This~~ temporal variation of the vertical distribution can be attributed to changes in the prevailing wind speed and direction throughout the day; as recorded by NOA's meteorological monitoring station at Thissio, the prevailing wind direction from 07:00UTC~~09:00LT~~ until 10:00UTC~~12:00LT~~ (LT=UTC+2 winter time and UTC+3 summer time) was from the south with speed from 1 to 4 $m \cdot s^{-1}$~~,m/s,~~ while easterly winds with speed reaching 10 $m \cdot s^{-1}$ started blowing at 11:00UTC,~~13:00 LT,~~ efficiently ventilating the Athens basin and removing the dust and atmospheric pollutants. As shown in previous works conducted in the area (e.g Fourtziou et al., 2017), wind speed below 3 $m \cdot s^{-1}$ favours the accumulation of pollutants.

The two cases, (ii) and (iii), present an elevated ~~two~~ aerosol layer~~s; a surface layer and an elevated one~~ extending up to 3 km between 10:00~~13:00~~ and 14:00UTC.~~17:00LT.~~ Lidar retrievals also show an elevated extinction layer in both cases, as discussed in section 3.3. However, the separation of the two layers could be an artifact which arises from the fact that the MAX-DOAS retrieval's response to a box-like distribution (e.g. a well developed planetary boundary layer - PBL) leads to slight oscillations around this box due to the a priori smoothing. Both cases are related to weak prevailing winds (<4 $m \cdot s^{-1}$), which favour the development of a vertically extended aerosol layer. ~~In case (iii) the near-surface aerosol load starts building up earlier than in case (ii).~~ The higher aerosol load in case (iii) is also corroborated by sun photometric measurements, which are presented and discussed in section 3.4.

LowA homogeneous surface aerosol layer with low levels of aerosol extinction (less than 0.1(0.2 km^{-1}) are maximum value) is present over the urban area throughout the whole day in case (iv). The highest valuesheight of the day (almost 0.14 km^{-1}) are displayed up toaerosol layer is around 800 m.a.s.l. Given that the NO_2 level, characteristic of anthropogenic pollution, is high during this day (Fig. 3), higher particle pollution levels would be expected.

3.3 MAX-DOAS aerosol extinction profiles evaluation

The BOREAS retrieved aerosol extinction profiles from the MAX-DOAS measurements at 477 nm, between 0.5 km (station's elevation) and 4 km height, are compared with the lidar aerosol extinction coefficient measurements at 532 nm, between 1 and 4 km height, for the selected case studies (Fig. 6). Representative morning and afternoon snapshots during each day have been chosen to be presented and discussed. The lack of morning profiles for some days is due to the absence of lidar data, thus, both morning and evening data is available only for cases (i) and (ii). The lidar profile presented used in each figure is the resultaverage of the mean lidar signal, averagedall profiles retrieved between the starting and the ending time of the corresponding presented MAX-DOAS profiles. The uncertainty in the lidar extinction profiles increases substantially for altitudes below 1000 m.a.s.l. due to the loss of overlap between the telescope field of view and the laser beam (Wandinger and Ansmann, 2002; Kim et al., 2008, Papayiannis et al., 2008); hence the lidar data for altitudes below 1000 m a.s.l. is not presented and only measurements above 1000 m a.s.l. are considered for the calculation of the correlation between the two instruments. Another point that has to be considered when comparing the results from the two instruments is that the lidar profiles are characterised by high vertical and temporal resolution and degradation to the sensitivity of the MAX-DOAS profiles is necessary in order to have a meaningful comparison to the MAX-DOAS data. According to the method described by Rodgers and Connor (2003), the degraded lidar profile x_f can be estimated by applying the equation

$$x_f = x_a + AK \cdot (x - x_a) \quad (3)$$

with x_a being the a priori profile used in the algorithm calculations, x the initial lidar profile and AK the averaging kernel from the BOREAS retrieval. The averaging kernel (Fig. 5) denotes the sensitivity of the retrieved profile to the true atmospheric profile for each layer and in fact it represents the smoothing of the true profile in the retrieval. The lidar profile, degraded to 50 m vertical resolution, represents the MAX-DOAS profile that would have been retrieved, if the true extinction profile was x. Last, but not least, the horizontal distance (13 km) between the two measurement sites and the different operation principles of the two instruments should be noted. The lidar system retrieves information from the air mass right above the measurement site, while MAX-DOAS probes air masses along the line of sight of the telescope pointing from the top of a hill towards the city centre; hence the retrieved aerosol profiles from the two instruments correspond to different air masses andcentre. Thus, some discrepancies are not expected to fully agree,, especially when the aerosol pollution is not horizontally homogeneous over the Athens basin. Thus,basin, and the comparison is mainly focused on a qualitative basis.

Each case is examined separately. Comparison information is given in the form of ~~and some~~ performance statistics - correlation coefficient (r), median lidar/MAX-DOAS ratio, root mean square error (RMSE) and~~,~~ fractional gross error (FGE) – and is ~~are~~ shown in Table 4.~~3.~~ This set of statistics has been chosen as suitable to provide a detailed view of the algorithm performance; it has been proposed (Morris et al., 2005) that a FGE less than or equal to 0.75 is a criterion to evaluate good performance of an algorithm, therefore, any FGE>0.75 is used as indicator of a relatively poor performance in this study. In order to perform the statisticals~~statistics~~ calculations we averaged the four MAX-DOAS profiles comprising each case. Thus, all performance statistics have been calculated using the temporally averaged MAX-DOAS profile for each case and the corresponding degraded lidar profile, so that both profiles are of the same temporal and vertical resolution. In all cases, 61 data points are used for the derivation of the statistics. The average smoothing and noise errors for the MAX-DOAS retrievals are given for each case study separately in Table 5. In all cases, the noise error ranges between about 1 and 5%. In cases (i)-mor, (ii) and (iii), the smoothing error is about 15%. The large smoothing errors in cases (i)-aft and (iv) are due to the very small extinction values at higher altitudes.

Case study (i) - 5 February 15

In case (i), the two instruments seem to be in excellent agreement~~,~~ in terms of ~~both extinction levels - up to 3 km - and~~ correlation, with a very high ~~when the morning measurements are considered; the median ratio is close to unity and the~~ correlation coefficient (r>0.95).~~is very high (r=0.98).~~ In the afternoon, a peak in aerosol extinction (~ 0.15 km^{-1}) between 1 and 1.5 km is captured by both instruments. The~~at 1.5 km is recorded by the MAX-DOAS in contrast to the lidar profile, which shows no elevated aerosol layer, leading to lower - still high - correlation (r=0.84). However, the~~ large discrepancy between the original and the degraded lidar profile is attributed to the fact that the AKs of the afternoon retrievals illustrate low sensitivity of the retrieved profile to the true atmospheric profile for altitudes up to 2.5 km (Fig. 5).

It should be mentioned that this is the only case in the present study, where high aerosol load is found in the upper levels (free troposphere) in the original lidar profiles due to transboundary transport of aerosols at higher altitudes. The fact that, at these altitudes, the MAX-DOAS only agrees with the degraded lidar profiles (which means after including the AK information) suggests more significant errors in the a priori aerosol profiles and the reduced capacity of the MAX-DOAS to capture the characteristic inhomogeneity at higher atmospheric layers during aerosol transport episodes. Nevertheless, an overall satisfactory performance of the algorithm is indicated for the morning measurements by the FGE (0.31).~~(0.6).~~

Case study (ii) - 9 July 15

The retrieved MAX-DOAS profiles agree quite well with the degraded lidar profiles; they both show an aerosol layer extending up to about 2.5 km and the correlation coefficient is very high (r ≈ 0.95),~~0.9),~~ during both morning and afternoon measurements. In the afternoon, however, the MAX-DOAS measurements result in higher extinction levels by almost 65%~~75%~~ compared to the degraded lidar profile. As shown in Fig. 6 (middle row panels), in this case MAX-DOAS tends to overestimate the lidar extinction levels mainly at higher altitudes, a fact that can be attributed to the smoothing effect of the retrieval procedure on the true profile; given that a MAX-DOAS profile algorithm cannot retrieve sharp edges, the underlying narrow high altitude enhancement in the afternoon propagates through the retrieval into a smoother and broader aerosol peak. The FGE, ranging from 0.35~~0.45~~ to 0.55, indicates a good performance of the algorithm.

Case study (iii) - 10 July 15

The two instruments seem to correlate very well (r=0.97). The MAX-DOAS coincides well with the aerosol extinction levels from the degraded lidar profile;~~lidar measurements, especially in the first 2 km;~~ the lidar to MAX-DOAS ratio is equal to 0.99.~~0.91.~~ Nevertheless, when the original lidar profile is considered, a clear discrepancy in the extinction levels is present; the lidar peak value (0.16 km^{-1}) is enhanced by a factor of two. The discrepancy between the original and the degraded lidar profile results from the low sensitivity of the averaging kernels for heights up to about 2 km (Fig. 5, case (iii)), which plays significant role in the degradation (smoothing) of the lidar retrieval. The RMSE is small (0.01 km^{-1}) and the low FGE (0.20)~~(0.23)~~ indicates good performance of the algorithm.

Case study (iv) - 4 April 16

The profiles resulting from both instruments display an aerosol layer extending from the lower atmospheric layers up to~~of about~~ 1.5 km height.~~deep.~~ The MAX-DOAS and degraded lidar profile shapes are very similar and highly correlated~~the correlation coefficient (r=0.90).(r=0.86) indicates a high correlation between the two instruments.~~ However, the MAX-DOAS underestimates by almost 30%~~50%~~ the lidar aerosol extinction (median lidar/MAX-DOAS ratio=1.58).~~ratio=1.8).~~ Although the correlation is high and the RMSE is small (0.02), the ~~relatively high~~ FGE (0.61)~~(0.77)~~ indicates a moderate~~poor~~ performance of the algorithm for the specific case. This FGE value, however, results from the high median ratio of the two profiles, which in turn results from the low extinction levels, since the absolute difference between the two profiles is not that large.

Overall, the correlation between lidar and MAX-DOAS measurements is very good (0.90~~(0.84~~ < r < 0.97)~~0.98)~~ in all cases and a good agreement in the profile shape and altitude of the peak extinction level is also observed. The failure of the MAX-DOAS to capture clearly distinguished aerosol layers is attributed to the smoothing effect due to the presence of a priori constraints during the retrieval procedure. The RMSE ranges from 0.01 to 0.04~~0.05~~ km-

[1] in all cases. A ~~satisfactory fractional gross error (<0.60), indicating a very good performance of the MAX-DOAS profiling algorithm, has been calculated in all cases with average or~~ high ~~extinction values. A higher~~ FGE (0.80)~~(0.77)~~ has been calculated only in the case of measurements at~~very~~ small relative azimuthal angles between the viewing direction and the Sun and in parallel presence of large particles.~~extinction levels.~~ Due to the different operation principles of each instrument (active/passive remote sensing), the different wavelengths and the different air masses probed by each instrument, a full agreement in the derived profiles would not be expected. In particular, the lidar profiles represent the aerosols which are directly over the measurement site, whereas the MAX-DOAS profiles are representative of the atmosphere at a distance of several kilometres along the line of sight of the instrument. Another conclusion arising from these four cases is that the MAX-DOAS fails to detect part of the urban aerosol pollution when the pollution levels are low (e.g. case(iv)) and also fails to capture the inhomogeneity at higher altitudes in case of aerosol transport episodes.

3.4 AOD evaluation

In the previous section, lidar measurements were used for the evaluation of the aerosol extinction profiles obtained from MAX-DOAS. However, a conclusive evaluation of MAX-DOAS aerosol optical depth (AOD) cannot be done strictly with lidar-derived AOD values mainly due to the lidar blind range (overlap height), resulting in AOD underestimation. Therefore, in this section, we focus on the comparison between the retrieved AOD~~In this section, we report on the comparison between the retrieved aerosol optical depth (AOD)~~ from MAX-DOAS measurements at 477 nm and from CIMEL measurements at two wavelengths (440 nm and 500 nm), during the aforementioned case studies (Fig. 7). For the MAX-DOAS AOD calculation, the missing values in the extinction coefficient profiles below 500 m are set to a constant value (equal to the retrieved value at 500 m). This assumes that the atmosphere is well-mixed below 500 m, which probably results in an underestimation of the calculated AOD in case of enhanced surface aerosol layer.~~7).~~ When looking at the figures, one should consider that the CIMEL AOD uncertainty is estimated to be approximately 0.01 for wavelengths > 400 nm (Eck et al., 1999). The MAX-DOAS AOD uncertainties are shown in the figures. The Ångström exponent, derived from the CIMEL measurements (400 - 870 nm), is also taken into account, as a qualitative indicator of aerosol particle size, in order to investigate the origin of the aerosols (natural-dust or anthropogenic sources) and the performance of the MAX-DOAS retrievals for different aerosol types and sizes. An overview of the comparison statistics (described in section 3.3), representative of the degree of agreement between MAX-DOAS and CIMEL measurements at 500 nm, is presented in Table 6.~~4.~~ The calculations were made on hourly basis to achieve uniform results regarding the air masses. Although this section 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. Nevertheless, as a complementary analysis, the comparison between lidar and MAX-DOAS AOD for the common altitude (1-4 km) - along with the corresponding uncertainties - is presented in Table 7.

Case study (i) - 5 February 15

The very small Ångström exponent, ranging between 0.05 and 0.13 throughout the day, indicates the dominance of coarse particles in the aerosol distribution. Given the cloud-free sky conditions and the potential for dust transport found for this day by the NOAA-HYSPLIT (Fig. 2), these particles are probably associated with the presence of dust in the atmosphere. TheAlthough the correlation between the two instruments is moderate (r=0.47) and(r=0.56), the calculated AOD levels from the MAX-DOAS measurements underestimate by about 20%agree very well with the CIMEL measurements (median ratio CIMEL/MAX-DOAS = 1.22).1.08). The daily averaged AOD values are 0.330.38 (± 0.02) and 0.39 (± 0.03) for MAX-DOAS and CIMEL, respectively, and are much higher than the climatological monthly average value (0.27 ± 0.03) for February in Athens, as reported in Gerasopoulos et al. (2011). The RMSE is 0.07, howeversmall (0.03) and the FGE, which is small (0.17)close to zero (0.09) implies excellent performance of the algorithm. The moderate correlation results may arise from the fact that CIMEL performs direct sun measurements, whereas MAX-DOAS measurements - and the subsequent AOD retrieval - are performed at a fixed azimuthal direction. Thus, the CIMEL measurements are highly affected by variations in the temporal and spatial distribution of the aerosols. The AOD from the lidar measurements at 10:0012:00 and 13:00UTC15:00LT (0.37 and 0.35, respectively) coincides well both with CIMEL and MAX-DOAS measurements.

Case study (ii) - 9 July 15

In this case, the large values of the Ångström exponent ($\alpha \geq 2$) are indicative of the presence of fine mode aerosols that are associated with urban pollution (Westphal and Toon, 1991, Eck et al., 1999, Gerasopoulos et al., 2011). The considerable levels of NO_2 measured during this day (Fig. 3), indicate the presence of anthropogenic pollution. The two instruments are again moderately correlated (r=0.67),(r=0.63), however if the afternoon measurements - after 16:00LT - are excluded, the correlation becomes very good (r=0.87).(r=0.86). The median ratio (0.85)(0.72) indicates that the MAX-DOAS overestimates the AOD levels measured by the CIMEL. However, the overestimation is more profound during the morning, while in the afternoon the MAX-DOAS slightly underestimates the measured AOD by about 20%,10%, a fact that can be attributed to inaccuracies based on the geometry of the light scattering in the radiative transfer calculation for the forward scattering geometry.direction. The daily averaged AOD values are 0.200.23 and 0.19 for MAX-DOAS and CIMEL, respectively. The small values of both the RMSE (0.07)(0.09) and the FGE (0.26)(0.35) are indicators of very good performance of the algorithm. The gaps in the CIMEL data in this case, as well as in case (iii), are probably due to saturation of the instrument. The lidar AOD in this case (0.21 at 10:00UTC13:00LT and 0.26 at 13:00UTC)16:00LT) agrees well with the CIMEL measurements at 500 nm, but is lower than the AOD from the MAX-DOAS; the difference is more considerable in the afternoon.

Case study (iii) - 10 July 15

In the third case study, the measurements from the two instruments seem to be in better agreement during morning hours. Overall, the MAX-DOAS ~~overestimates the~~ AOD levels coincide well with~~from~~ CIMEL (median CIMEL/MAX-DOAS ratio is 0.95),~~by 20%,~~ however the underestimation due to light scattering geometry ~~is about 25%~~ after 13:00UTC is about 35%.~~(16:00LT).~~ The moderate results with respect to correlation (r=0.53)~~(r=0.55)~~ are due to the large discrepancy between the two instruments during the afternoon. If only the measurements until 13:00UTC~~16:00LT~~ are considered, the correlation is considerably improved (r=0.75). Despite the non-satisfactory correlation, the calculated FGE (0.28)~~(0.32)~~ indicates a very good performance of the algorithm. The lidar-derived AOD in the afternoon is higher than~~coincides very well with~~ the MAX-DOAS measurement. Unfortunately, no CIMEL or lidar data are available around noon in order to validate the aerosol plume captured by MAX-DOAS. It should be noted, though, that case studies (ii) and (iii) (both summer days in July) exhibit the same diurnal pattern; lower values in the morning, steadily increasing throughout the day and then slightly declining in the afternoon. A similar diurnal AOD pattern was found for summer in Athens by Gerasopoulos et al., 2011 and this pattern has been associated with local urban or industrial sources (Smirnov et al., 2002).

Case study (iv) - 4 April 16

The Ångstöm exponent in this case (α≈1) indicates the presence of coarse aerosols (radii≥0.5 μm) in the atmosphere (Westphal and Toon, 1991, Eck et al., 1999). The NOAA HYSPLIT backtrajectories show the potential for African dust transport to Athens, however, at higher altitudes of up to 5 km. Nevertheless, despite the presence of coarse particles, the AOD levels are low; the daily averaged AOD values are 0.13~~0.15~~ and 0.19 for MAX-DOAS and CIMEL, respectively. The MAX-DOAS underestimates the AOD with respect to CIMEL by about 25%~~15%~~ (and by 50%~~30%~~ if only the afternoon measurements are considered), the RMSE is 0.07~~0.05~~ and the FGE is 0.40,~~0.27,~~ yet it seems that the two measurement techniques are not correlated (r=-0.42).~~(r=-0.44).~~ However, the comparison in terms of correlation, results in better outputs if only the morning measurements are considered; in this case the correlation coefficient is 0.75.~~0.80.~~ It seems that MAX-DOAS, in this case, fails to detect the accumulation of coarse particles. Nevertheless, the AOD lidar measurements ~~(AOD=0.15) in the afternoon~~ agree very well with the MAX-DOAS measurements (Table 7).~~AOD (0.14).~~

Overall, a systematic underestimation of the AOD, by 20~~10~~ to 35%,~~30%,~~ by the MAX-DOAS is observed in the afternoon measurements, when the relative azimuthal angle between the MAX-DOAS viewing direction and the Sun is small. Better agreement is achieved at large relative azimuthal angles in the morning. This finding has also been reported by Frieß et al. (2016) when comparing different retrieval algorithms with sun photometer measurements. Considering that i) the~~The~~ sun-photometer is located downtown (150 m a.sl.), at lower altitude than the MAX-DOAS (527 m a.s.l.) and~~,~~ thus ~~it is~~ more sensitive to aerosols in the lower troposphere and ii) the absence of real measurements from MAX-DOAS for altitudes below 500 m a.s.l., 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 0.03~~0.05~~ with standard deviation 0.08.~~0.09.~~ Furthermore, CIMEL is a direct sun photometer, which means that in each measurement different air masses are detected, while the MAX-DOAS always points at the same direction; this operational difference is reflected in the non-satisfactory correlation. When fixed values of SSA and asymmetry factor (instead of AERONET data) are used by BOREAS, it seems that MAX-DOAS fails to detect accumulated coarse particles (e.g. case (iv)), leading to underestimation in case of small Ångström exponent values (< 1). Frieß~~Friess~~ et al. (2016)~~(2017)~~ have also come to this conclusion during the CINDI-2 campaign. However, the underestimation could also be attributed to the high altitude aerosol layer detected by the lidar (Fig. 6); the MAX-DOAS' sensitivity at higher altitudes is low and the calculated AOD is limited up to 4 km, while the AOD from CIMEL refers to the total atmospheric column. In addition, the presence of aerosol layers above 4 km that could contribute to the AOD was examined by using the lidar signals and no significant aerosol load was observed above this height.~~, thus it records lower AOD levels.~~ It should also be noted that the standard AERONET version 2 algorithm uses an NO_2 climatology with a spatial resolution much coarser~~higher~~ than the area of the~~Athens~~ city of Athens~~area~~ (Giles et al., 2019), hence in certain cases the difference between the MAX-DOAS and the higher CIMEL AOD levels at 440 nm could result from additional NO_2 content in the atmosphere, which is the case of highly polluted days. The lidar-derived AOD levels coincide well with the MAX-DOAS ~~measurements in all cases, apart from the case (ii), in which MAX-DOAS overestimates both CIMEL and lidar~~ measurements.

4. Summary and conclusions

An assessment of the retrieval of aerosol extinction profiles and AOD from MAX-DOAS measurements is presented for the first time for the urban environment of Athens. The profiling results are compared to lidar extinction profiles and to AODs obtained from sun photometric measurements. The intercomparison results are very promising, showing that the MAX-DOAS measurements provide a good estimation of the aerosol vertical profile over Athens. Although this intercomparison is of great importance for the validation of the MAX-DOAS retrieval, the different operation, characteristics and measurement principles of each instrument, in addition to some comparison restrictions, have to be considered.

Regarding the spatial characteristics, (i) the measurements with the MAX-DOAS technique represent an area that includes the AERONET and the lidar locations, but it is not limited to them. Regarding the vertical aerosol information, (ii) the MAX-DOAS retrievals are representative for 500-4000 m a.s.l., while the lidar profiles are valid for altitudes higher than 1000 m above the station and finally (iii) the AOD AERONET measurements describe the columnar aerosol properties representative of an area ranging from few up to 10 km radius above the Athens area, depending on solar elevation. Also, (iv)~~(v)~~ the sun-photometer AOD observations probe the extinction in the full atmospheric column while MAX-DOAS

retrievals are sensitive only to the lowest kilometers, leading to differences in the presence of aerosol layers at altitudes above 4 km. Nevertheless, despite the comparison restrictions and the differences of the three instruments, the comparison of the retrieved profiles and the AODs shows that the MAX-DOAS measurements bode well for the future of aerosol measurements and they are able to provide a good estimation of the aerosol vertical distribution over Athens.

The vertical profiles retrieved by BOREAS profiling algorithm applied to the MAX-DOAS measurements are qualitatively in good agreement with the lidar profiles smoothed with the MAX-DOAS averaging kernels; there is good agreement in aerosol layer shape and aerosol extinction levels, except in ~~most of the~~ cases of inhomogeneity at higher altitudes, characteristic of aerosol dust transport episodes.~~.~~ Very good correlation ($r > 0.90$)~~0.84)~~ was found in all cases. A satisfactory fractional gross error (0.20~~(0.25~~ < FGE < 0.54)~~0.60)~~ has been calculated in all cases with fine aerosol particles (urban pollution),~~average or high extinction levels,~~ indicating a good performance of the BOREAS profiling algorithm in these cases. In some cases, the observed underestimation of the aerosol extinction (by 20~~10~~ to 35%)~~30%)~~ by the MAX-DOAS at small relative azimuth angles can be attributed to the geometry of Mie scattering in relation to the location and viewing geometry of MAX-DOAS, resulting in MAX-DOAS' failure to detect part of the urban aerosol pollution. Overall, the agreement between the two instruments is encouraging, especially when considering the different nature of each technique and the different instrument locations, suggesting that the MAX-DOAS can accurately enough represent the aerosol vertical distribution.

The MAX-DOAS retrieved AODs show satisfactory agreement with the sun photometric measurements, in terms of AOD levels. The MAX-DOAS underestimates the AOD in the presence of coarse particles; CIMEL/MAX-DOAS ratio > 1 coincides with Ångström exponent values < 1. A systematic underestimation by MAX-DOAS is observed in the afternoon measurements due to MAX-DOAS' viewing geometry. Overall, the MAX-DOAS can be considered as an effective mean for measuring the aerosol levels in Athens; the average AOD difference of all measurements between the two instruments is ~~0.05.~~0.03. It is important to note that in Athens, a highly populated and polluted area, horizontal gradients, especially in anthropogenic aerosols, are very likely to occur, resulting in different air masses detected by each instrument and subsequently in discrepancies between MAX-DOAS and CIMEL measurements.

This intercomparison is of great importance for the validation of the MAX-DOAS retrieval. Despite the already mentioned limitations due to different operation, characteristics and measurement principles of each instrument,~~retrieval, despite the different operation, characteristics and measurement principles of each instrument. While evaluating the MAX-DOAS performance in tracking aerosols (AOD and vertical distribution) the following restrictions should be kept in mind: (i) the measurements with the MAX-DOAS technique~~

~~represent an area that includes the AERONET and the lidar locations, but it is not limited to them. Regarding the vertical aerosol information, (ii) the MAX-DOAS retrievals are representative for 500-4000 m a.s.l., while the lidar profiles are valid for altitudes higher than 1000 m above the station and finally (iii) the AOD AERONET measurements describe the columnar aerosol properties representative of an area ranging from few up to 10 km radius above the Athens area, depending on solar elevation. Also, (v) the sun-photometer AOD observations probe the extinction in the full atmospheric column while MAX-DOAS retrievals are sensitive only to the lowest kilometers, leading to differences in the presence of aerosol layers at altitudes above 4 km.~~

~~Despite the mentioned limitations,~~ this work demonstrates that the MAX-DOAS measurements in Athens and the BOREAS algorithm can provide a good estimation of the aerosol vertical structure of the urban atmosphere, on a continuous and long-term basis, offering a reliable data set for scientific studies. There is certainly more work to be conducted in future studies in order to understand the sensitivity of the MAX-DOAS aerosol measurements based on different aspects of urban pollution evolution and long range transported aerosols.

Data availability

All data sets used and produced for the purposes of this work are freely available and can be requested from the corresponding author.

Author contribution

MG, PK and AR conceived the presented idea. MG performed the analysis and prepared the manuscript. TB developed the profile retrieval algorithm and provided guidance to MG on the algorithm calculations and parameterization. AR provided guidance to MG on MAX-DOAS data retrieval and profile calculations. MG designed the figures with support from PK and TB. AR and SK advised MG on the results interpretation. PK, AP and MM provided the lidar data. VA provided the sun-photometer measurements. AT made comments on the sun-photometer/max-doas comparison. AR, EG, SK, NM and MV provided critical feedback. All authors provided comments that helped shape the manuscript.

Competing interests

Andreas Richter and Vassilis Amiridis are members of the editorial board of the journal.

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Figure 1: Measurement sites and MAX-DOAS viewing direction (S). The distances between instruments are: MAX-DOAS - sun-photometer (CIMEL) 16 km and MAX-DOAS - lidar 13 km.

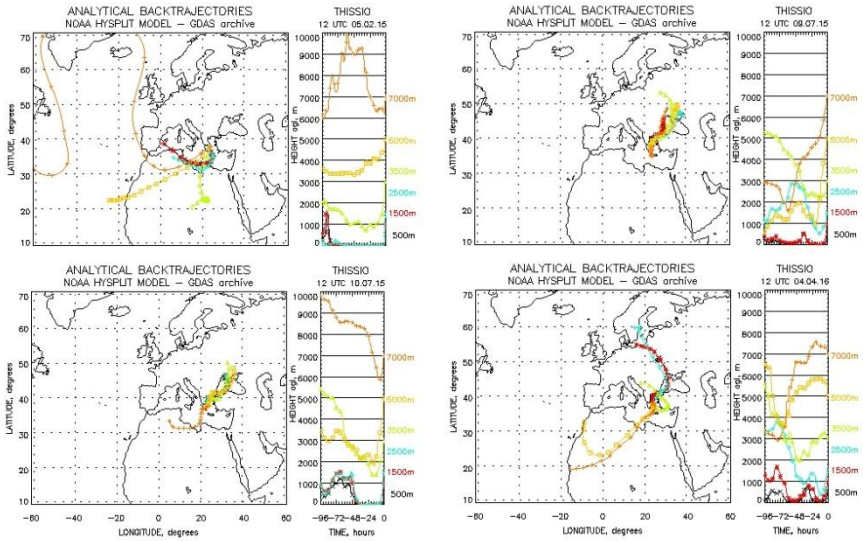
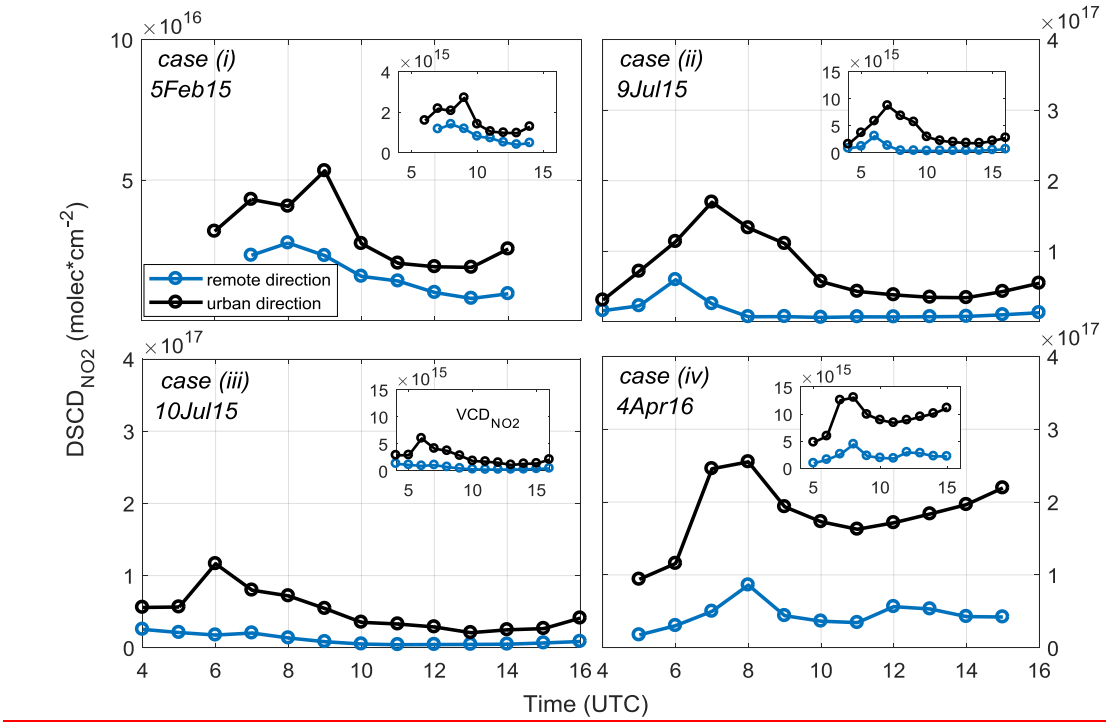


Figure 2: Analytical backtrajectories for Athens as derived from NOAA-HYSPLIT model for the case studies (i) 5Feb15 (top left panel), (ii) 9Jul15(top right panel, (iii) 10Jul15 (bottom left panel) and (iv) 4Apr16 (bottom right panel).



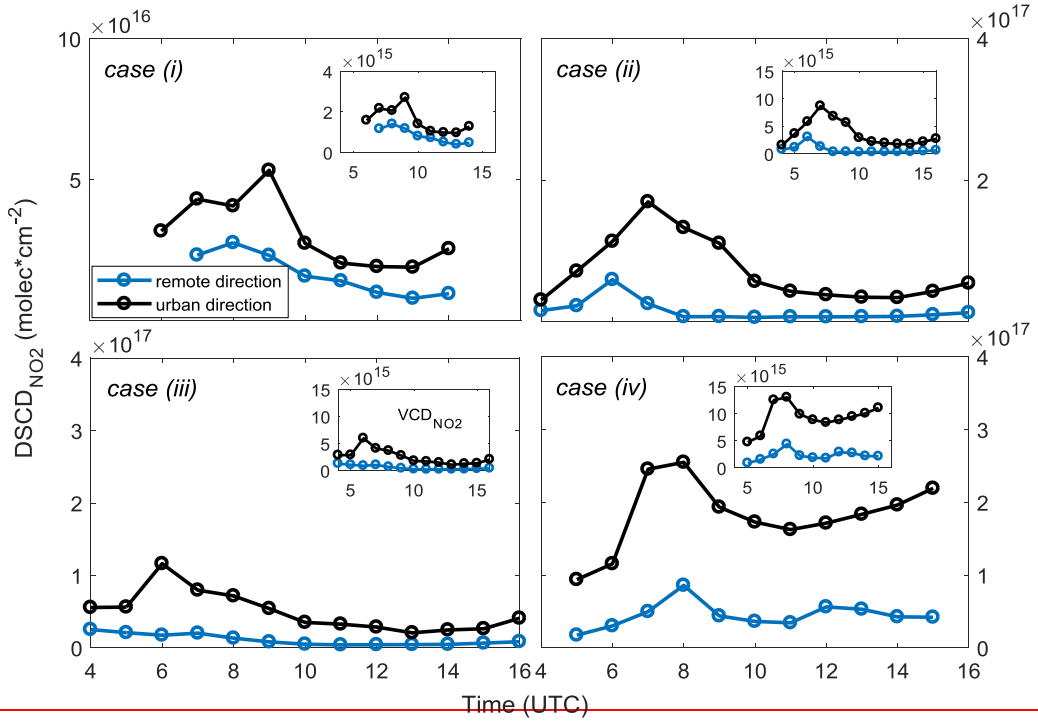
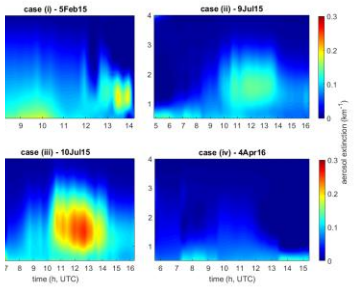
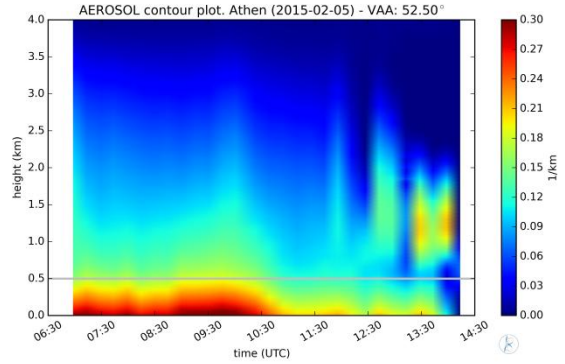
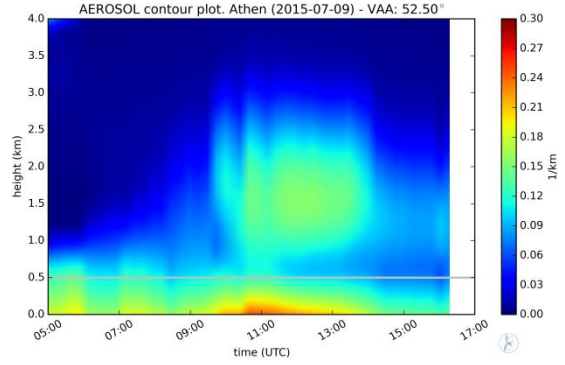


Figure 3: Tropospheric ~~SCD_{NO2}~~ retrievals of diurnal SCD_{NO2} (elevation angle +1°) from MAX-DOAS measurements for the four selected cases studies. The blue and the black curves correspond to the remote (W) and the urban (S) viewing direction, respectively. In the internal panels the corresponding tropospheric VCD_{NO2} are also shown. Please consider the different scale used in case (i).


case (i) - 5Feb15
case (ii) - 9Jul15
case (iii) - 10Jul15
case (iv) - 4Apr16
time (h, UTC)
time (h, UTC)
aerosol extinction (km⁻¹)


AEROSOL contour plot. Athen (2015-02-05) - VAA: 52.50°
height (km)
time (UTC)
1/km


AEROSOL contour plot. Athen (2015-07-09) - VAA: 52.50°
height (km)
time (UTC)
1/km

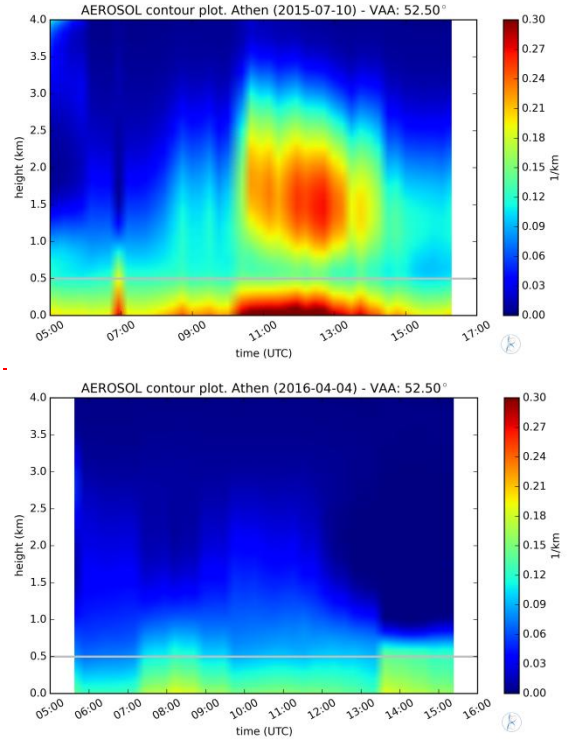


Figure 4: MAX-DOAS retrieved aerosol extinction vertical distributions (from instrument's height up to 4 km a.s.l.) for the four case studies over the urban area (S). ~~for case studies (i) (top left panel), (ii) (top right panel), (iii) (bottom left panel) and (iv) (bottom right panel).~~ The spatial and vertical resolution of the retrievals is 50 m and 15 min, respectively. ~~grey line depicts the station's height.~~

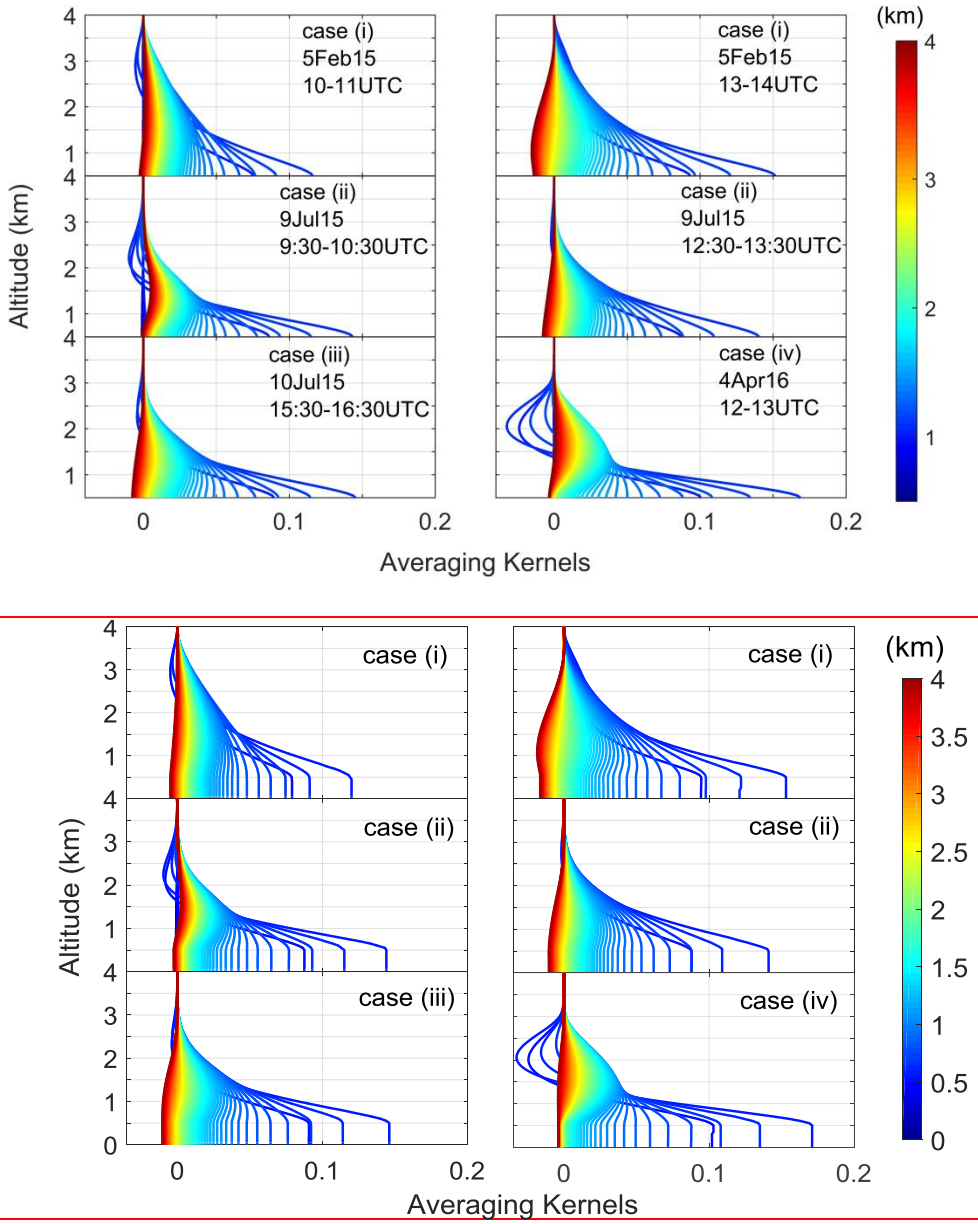
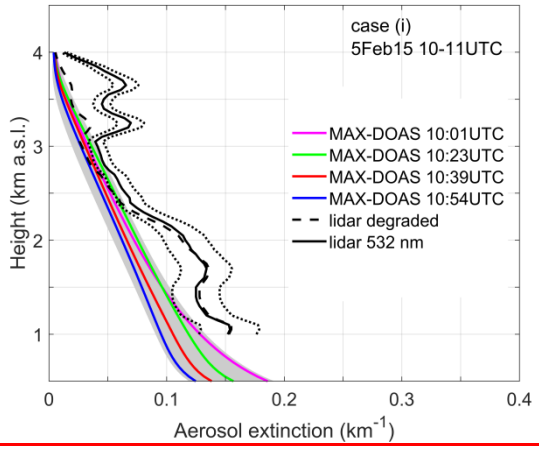
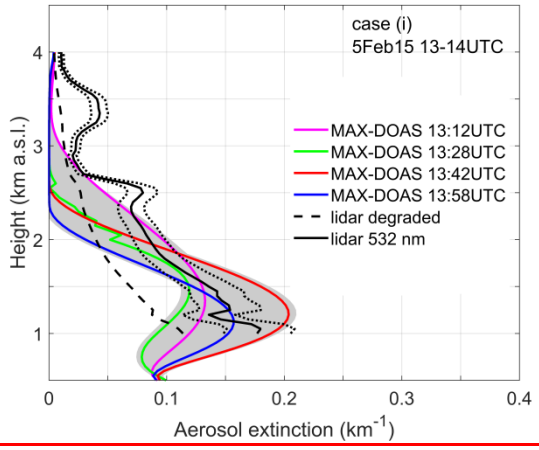
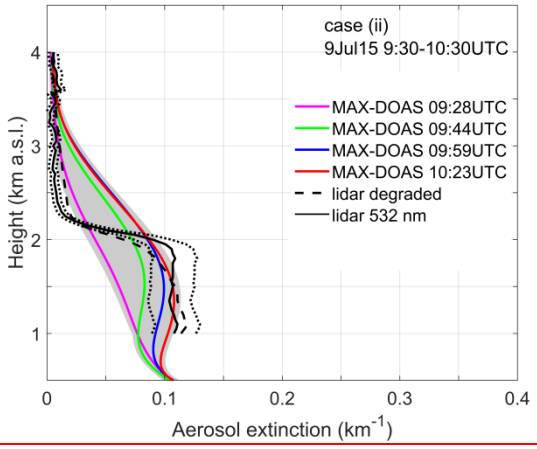
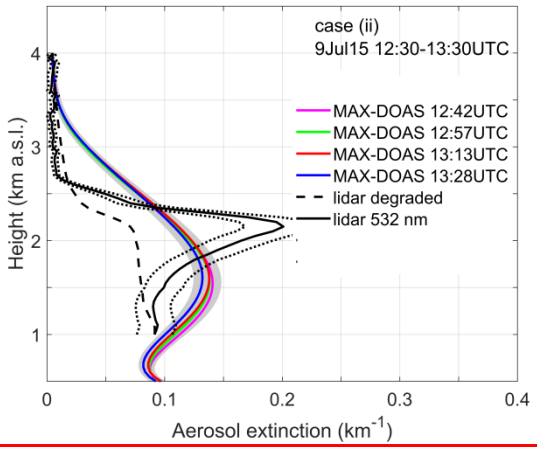


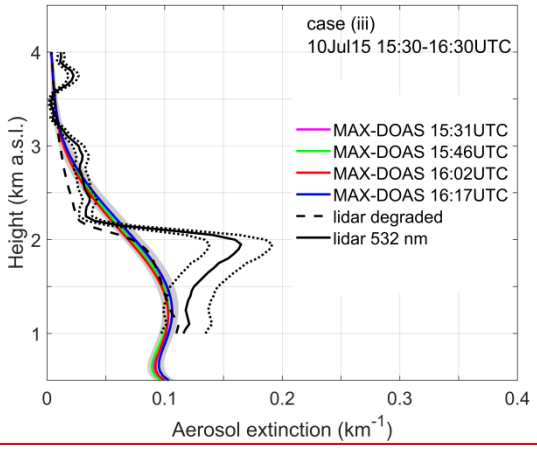
Figure 5: Averaging kernels of the aerosol retrievals for the four case studies. For cases (i) and (ii), the left and right panel corresponds to the morning and afternoon profiles, respectively. The colour bar represents the height of the atmospheric layers.

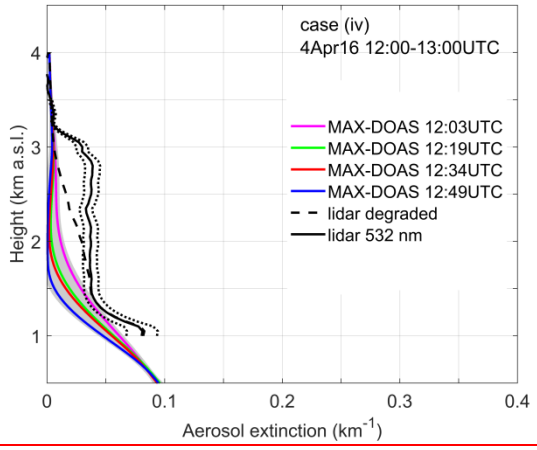

case (i)
5Feb15 10-11UTC
MAX-DOAS 10:01UTC
MAX-DOAS 10:23UTC
MAX-DOAS 10:39UTC
MAX-DOAS 10:54UTC
lidar degraded
lidar 532 nm
Height (km a.s.l.)
Aerosol extinction (km-1)

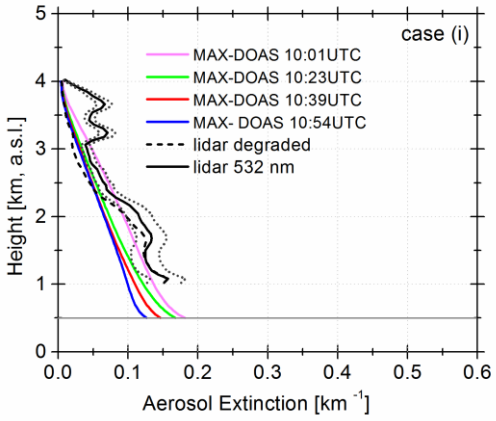

case (i)
5Feb15 13-14UTC
MAX-DOAS 13:12UTC
MAX-DOAS 13:28UTC
MAX-DOAS 13:42UTC
MAX-DOAS 13:58UTC
lidar degraded
lidar 532 nm
Height (km a.s.l.)
Aerosol extinction (km-1)

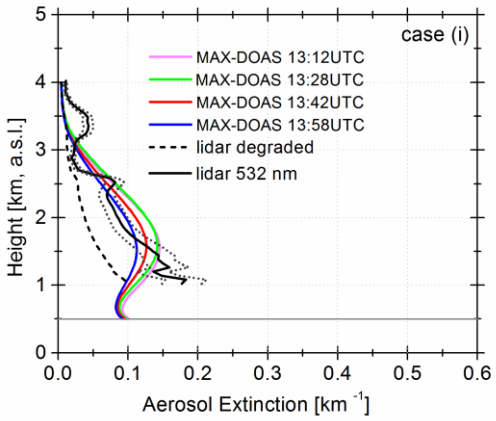

case (ii)
9Jul15 9:30-10:30UTC
MAX-DOAS 09:28UTC
MAX-DOAS 09:44UTC
MAX-DOAS 09:59UTC
MAX-DOAS 10:23UTC
lidar degraded
lidar 532 nm
Height (km a.s.l.)
Aerosol extinction (km-1)

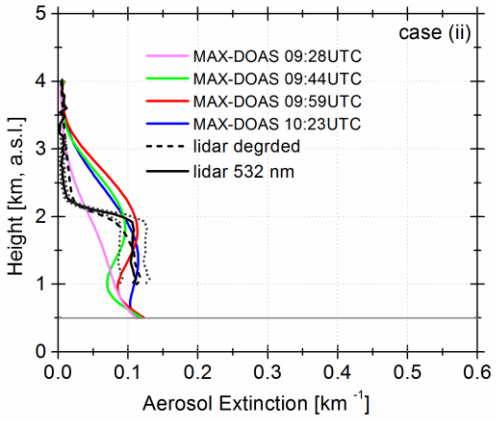

case (ii)
9Jul15 12:30-13:30UTC
MAX-DOAS 12:42UTC
MAX-DOAS 12:57UTC
MAX-DOAS 13:13UTC
MAX-DOAS 13:28UTC
lidar degraded
lidar 532 nm
Height (km a.s.l.)
Aerosol extinction (km-1)

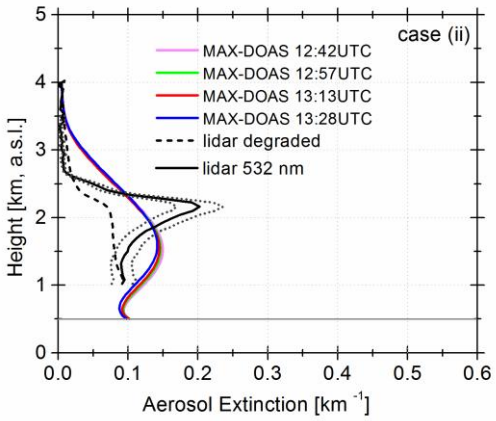

case (iii)
10Jul15 15:30-16:30UTC
MAX-DOAS 15:31UTC
MAX-DOAS 15:46UTC
MAX-DOAS 16:02UTC
MAX-DOAS 16:17UTC
lidar degraded
lidar 532 nm
Height (km a.s.l.)
Aerosol extinction (km-1)


case (iv)
4Apr16 12:00-13:00UTC
MAX-DOAS 12:03UTC
MAX-DOAS 12:19UTC
MAX-DOAS 12:34UTC
MAX-DOAS 12:49UTC
lidar degraded
lidar 532 nm
Height (km a.s.l.)
Aerosol extinction (km-1)


case (i)
MAX-DOAS 10:01UTC
MAX-DOAS 10:23UTC
MAX-DOAS 10:39UTC
MAX- DOAS 10:54UTC
lidar degraded
lidar 532 nm
Height [km, a.s.l.]
Aerosol Extinction [km -1]
0
1
2
3
4
5
0.0
0.1
0.2
0.3
0.4
0.5
0.6


case (i)
MAX-DOAS 13:12UTC
MAX-DOAS 13:28UTC
MAX-DOAS 13:42UTC
MAX-DOAS 13:58UTC
lidar degraded
lidar 532 nm
Height [km, a.s.l.]
Aerosol Extinction [km -1]
0
1
2
3
4
5
0.0
0.1
0.2
0.3
0.4
0.5
0.6


case (ii)
MAX-DOAS 09:28UTC
MAX-DOAS 09:44UTC
MAX-DOAS 09:59UTC
MAX-DOAS 10:23UTC
lidar degrded
lidar 532 nm
Height [km, a.s.l.]
Aerosol Extinction [km -1]
0
1
2
3
4
5
0.0
0.1
0.2
0.3
0.4
0.5
0.6


case (ii)
MAX-DOAS 12:42UTC
MAX-DOAS 12:57UTC
MAX-DOAS 13:13UTC
MAX-DOAS 13:28UTC
lidar degraded
lidar 532 nm
Height [km, a.s.l.]
Aerosol Extinction [km -1]
0
1
2
3
4
5
0.0
0.1
0.2
0.3
0.4
0.5
0.6

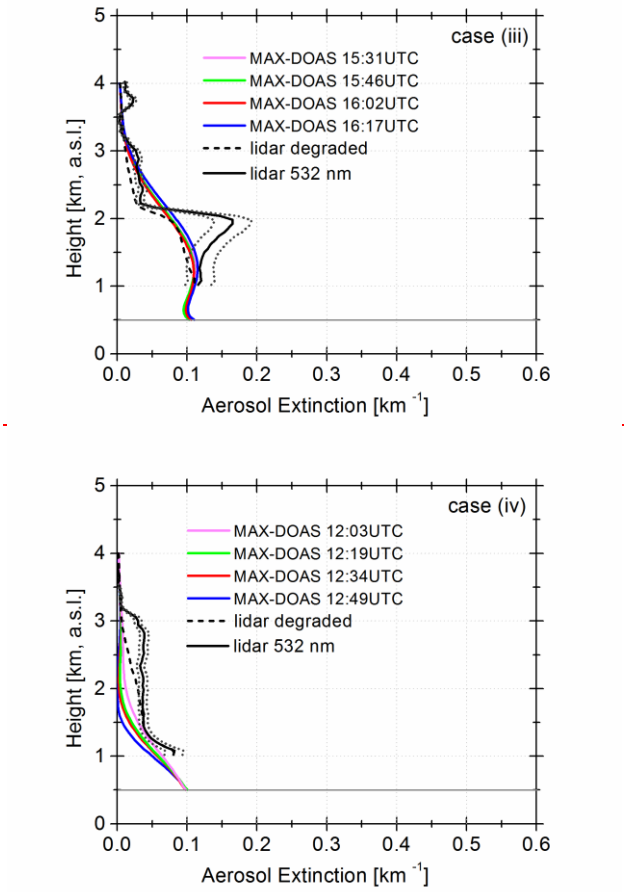
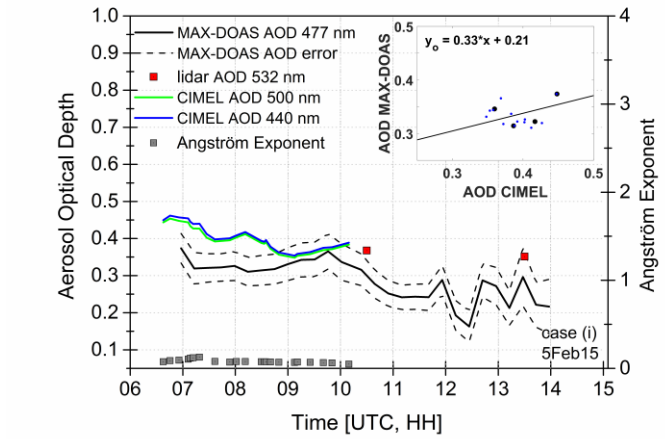
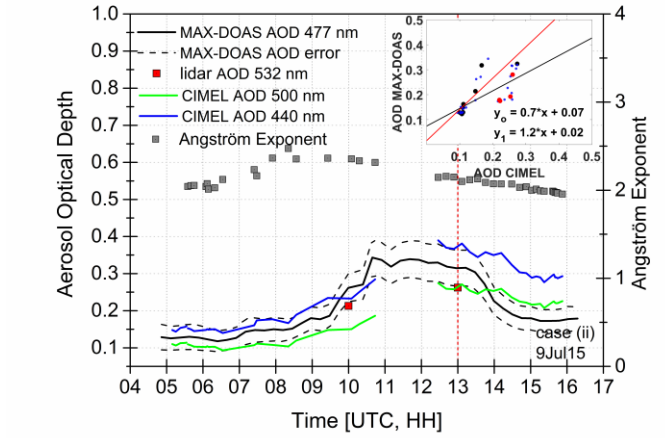
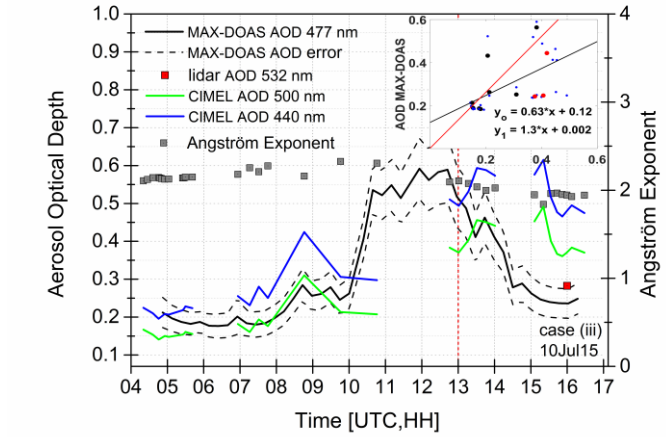
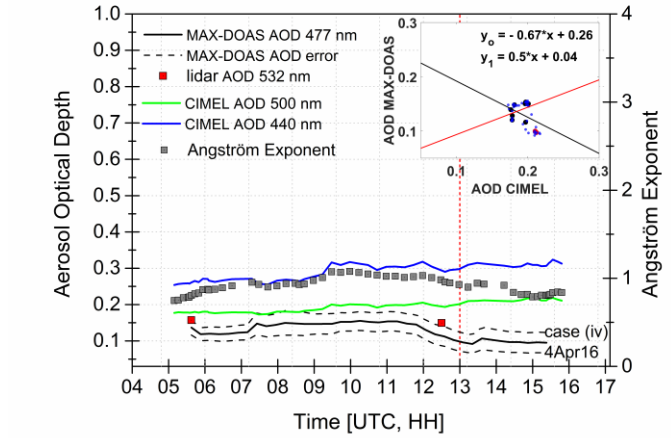


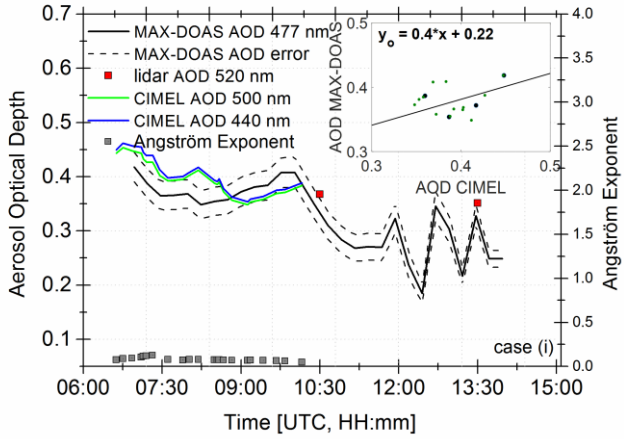
Figure 6: Comparison of retrieved MAX-DOAS aerosol extinction profiles at 477 nm (multicoloured curves), lidar aerosol extinction coefficient vertical profile at 532 nm (black curve) and the corresponding degraded lidar profile (dashed black curve) for the selected case studies. The lidar profile used in each case is the average profile retrieved between the starting and the ending time of the MAX-DOAS retrievals and the light dashed black curves are the lidar-derived aerosol extinction uncertainty obtained by the lidar assumption of 50 ± 20 sr. ~~.~~ The grey shaded area represents the corresponding ~~line depicts the elevation of~~ MAX-DOAS uncertainty.~~station at 500 m.a.s.l.~~

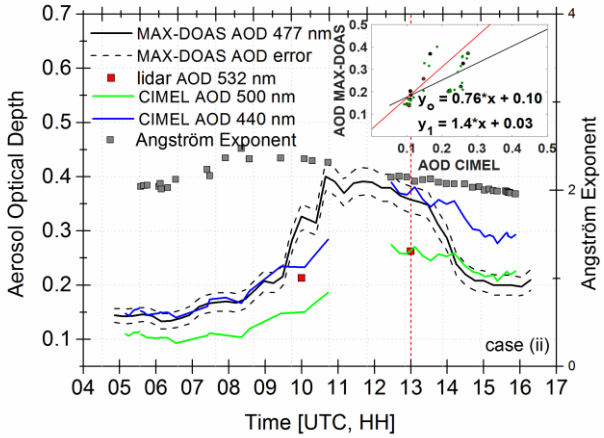

MAX-DOAS AOD 477 nm
MAX-DOAS AOD error
lidar AOD 532 nm
CIMEL AOD 500 nm
CIMEL AOD 440 nm
Angström Exponent
y_o = 0.33*x + 0.21
AOD MAX-DOAS
AOD CIMEL
Aerosol Optical Depth
Angström Exponent
Time [UTC, HH]
case (i)
5Feb15

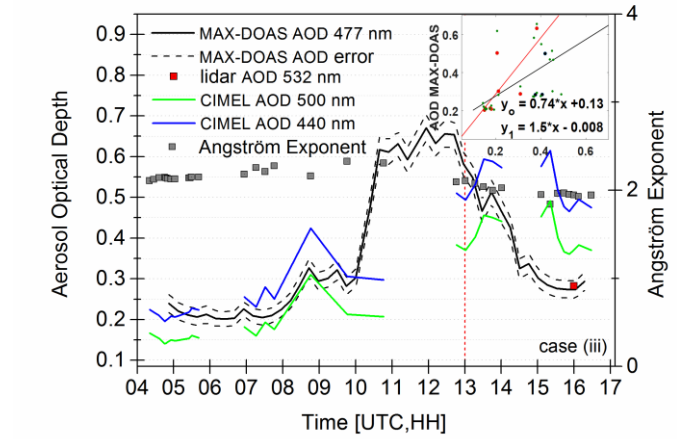

MAX-DOAS AOD 477 nm
MAX-DOAS AOD error
lidar AOD 532 nm
CIMEL AOD 500 nm
CIMEL AOD 440 nm
Angström Exponent
y_o = 0.7*x + 0.07
y_1 = 1.2*x + 0.02
AOD MAX-DOAS
AOD CIMEL
Aerosol Optical Depth
Angström Exponent
Time [UTC, HH]
case (ii)
9Jul15


MAX-DOAS AOD 477 nm
MAX-DOAS AOD error
lidar AOD 532 nm
CIMEL AOD 500 nm
CIMEL AOD 440 nm
Angström Exponent
AOD MAX-DOAS
y_o = 0.63*x + 0.12
y_1 = 1.3*x + 0.002
case (iii)
10Jul15
Aerosol Optical Depth
Angström Exponent
Time [UTC,HH]


MAX-DOAS AOD 477 nm
MAX-DOAS AOD error
lidar AOD 532 nm
CIMEL AOD 500 nm
CIMEL AOD 440 nm
Angström Exponent
AOD MAX-DOAS
y_o = - 0.67*x + 0.26
y_1 = 0.5*x + 0.04
AOD CIMEL
case (iv)
4Apr16
Aerosol Optical Depth
Angström Exponent
Time [UTC, HH]


MAX-DOAS AOD 477 nm
MAX-DOAS AOD error
lidar AOD 520 nm
CIMEL AOD 500 nm
CIMEL AOD 440 nm
Angström Exponent
y_o = 0.4*x + 0.22
AOD MAX-DOAS
AOD CIMEL
case (i)
Aerosol Optical Depth
Angström Exponent
Time [UTC, HH:mm]
06:00
07:30
09:00
10:30
12:00
13:30
15:00


MAX-DOAS AOD 477 nm
MAX-DOAS AOD error
lidar AOD 532 nm
CIMEL AOD 500 nm
CIMEL AOD 440 nm
Angström Exponent
y_o = 0.76*x + 0.10
y_1 = 1.4*x + 0.03
AOD MAX-DOAS
AOD CIMEL
case (ii)
Aerosol Optical Depth
Angström Exponent
Time [UTC, HH]



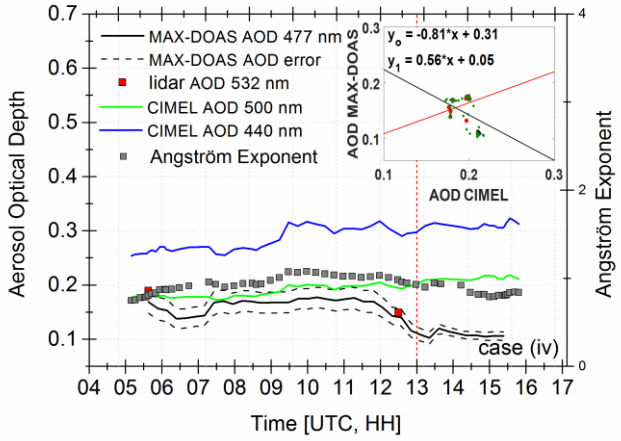


Figure 7: AOD as derived from MAX-DOAS (black curve) and CIMEL at 440 nm and 500 nm (green and blue curve, respectively). The grey and the red square markers represent the Angström exponent derived from 400 and 870 nm and the lidar derived AOD, respectively. The dashed black curves represent the MAX-DOAS AOD uncertainties. The scatter plots between hourly AOD calculated from MAX-DOAS measurements and hourly AOD from CIMEL at 500nm are shown in the internal panels; the red points correspond to measurements after 13:00UTC.~~until 16:00LT.~~ Accordingly, y_0 is the linear regression equation with all the data points included and y_1 is the linear regression equation when the data points after 13:00UTC~~16:00LT~~ have been excluded. The smaller bluegreen points are the raw data points. The vertical red dashed line separates the measurement data before and after 13:00UTC.~~16:00LT.~~

Table 1: Instruments and data products used in the present study.

Instrument	Location	Institute	Products
MAX-DOAS	Penteli National Observatory of Athens (38.05°N, 23.86°E, 527 m a.s.l)	BREDOM network, Institute of Environmental Physics and Remote Sensing, University of Bremen	SCD_{NO2}, VCD_{NO2}, aerosol extinction profile, AOD
EOLE-LIDAR	Zografou (37.97°N, 23.79°E, 212 m a.s.l.)	National Technical University of Athens, Laser Remote Sensing Laboratory (NTUA-LRSU)	Aerosol backscatter profile, aerosol extinction profile, columnar AOD

CIMEL Sun-Sky Radiometer	Thissio (37.96°N, 23.72°E, 150 m a.s.l.)	National Observatory of Athens, Institute for Astronomy, Astrophysics Space Application & Remote Sensing (NOA-IAASARS)	AOD, Inversion data products (ssa, asymmetry factor, refractive index, phase function, size distribution)

Table 2. Information about the selected case studies.

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

Table 3. Settings used for the BOREAS retrieval. The mean daily value of each parameter (ω and g retrieved from AERONET) is mentioned for cases (i), (ii) and (iii). The mean ~~Next year's~~ monthly values ~~mean~~ of ω and g (provided from AERONET for April 2017) were used for case (iv), due to unavailable AERONET daily data around this date.

	case (i)	case (ii)	case (iii)	case (iv)
Surface albedo	0.15	0.15	0.15	0.15
Single scattering albedo (ω)	0.92	0.96	0.93	0.91
Asymmetry factor (g)	0.78	0.65	0.68	0.68
Tikhonov parameter	20	20	20	20

Table 4.~~3.~~ Quantitative performance statistics of MAX-DOAS aerosol extinction calculations (BOREAS algorithm) compared to lidar measurements.

Performance Measure	case (i)-mor	case (i)-aft	case (ii)-mor	case (ii)-aft	case (iii)	case (iv)
r	0.97~~0.98~~	0.96~~0.84~~	0.92~~0.86~~	0.95	0.97	0.90~~0.86~~
median ratio (lidar/MAXDOAS)	1.37~~0.92~~	1.11~~0.45~~	0.91~~0.84~~	0.60~~0.57~~	0.99~~0.91~~	1.58~~1.80~~
RMSE (km^{-1})	0.03~~0.02~~	0.03~~0.05~~	0.02	0.04	0.01	0.02
FGE	0.31~~0.27~~	0.80~~0.60~~	0.37~~0.44~~	0.54~~0.55~~	0.20~~0.23~~	0.61~~0.77~~

Table 5. The MAX-DOAS average smoothing and noise errors (%) for each case study.

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

Table 6.~~Table 4.~~ Quantitative performance statistics of MAX-DOAS AOD calculations (BOREAS algorithm) at 477 nm compared to CIMEL measurements at 500 nm .

Performance	case (i)	case (ii)	case (iii)	case (iv)

Measure				
r	0.470.56	0.670.63	0.530.55	-0.42-0.44
median ratio (CIMEL/MAXDOAS)	1.221.08	0.850.72	0.950.83	1.371.19
RMSE	0.030.07	0.060.09	0.110.13	0.070.05
FGE	0.170.09	0.260.35	0.280.32	0.400.27

Table 7. MAX-DOAS (477 nm) and lidar (532 nm) AOD calculations for the atmospheric layer 1-4 km.

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