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.

<u>Retrieval shortcomings</u>

<u>1. Observation geometry</u>

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 troposphericbromine 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. 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 the figures in the revised manuscript.

- 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) 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.

- 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 <u>to exclude the retrievals below 500 m in the revised manuscript</u>. The updated Fig. 4 is attached at the end of this document.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, along with their comparison with the sun-photometer AOD.

<u>2. Standard atmosphere</u>

The profiles used for T and p affect the O_4 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).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.

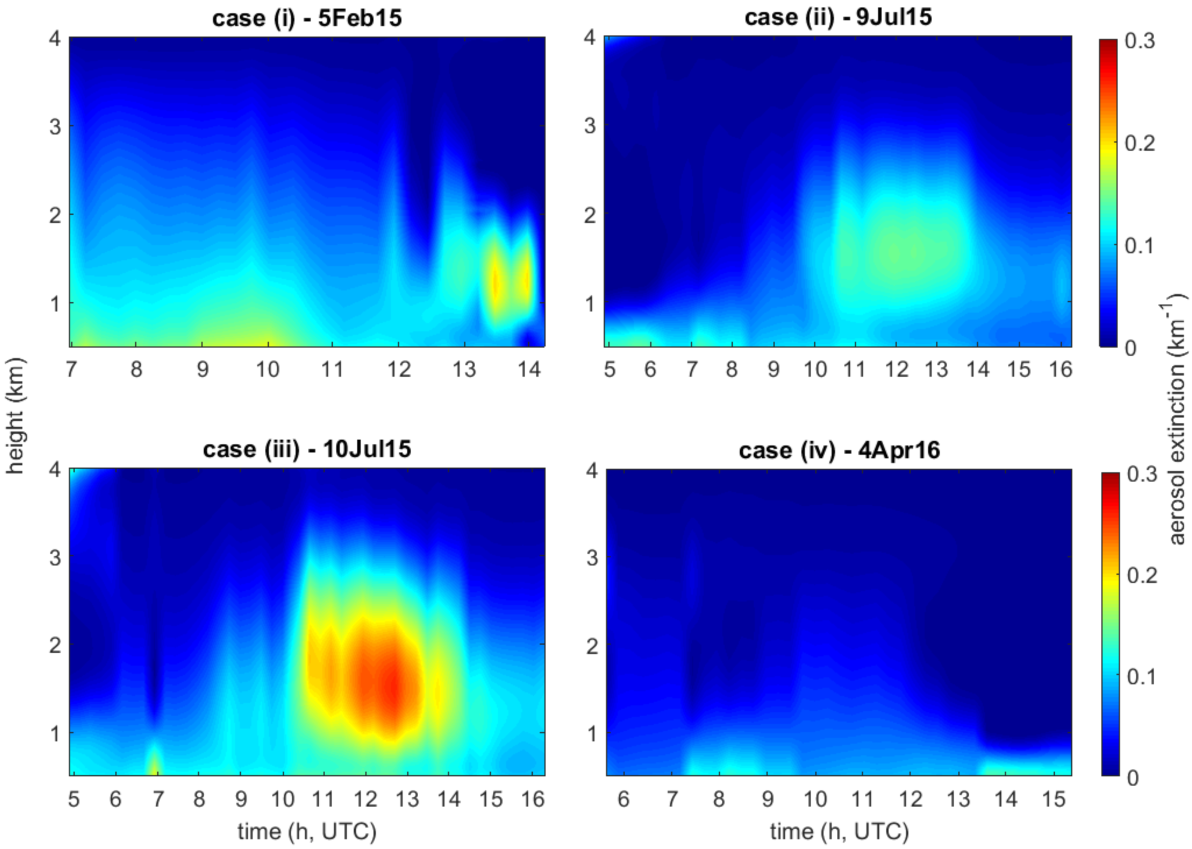

Figure 4: MAX-DOAS retrieved aerosol extinction vertical distributions (from instrument's height up to 4 km a.s.l.) for the four case studies over the urban area (S). The spatial and vertical resolution of the retrievals is 50 m and 15 min, respectively.