Response to Referee #2

We would like to thank the reviewer for carefully reading the manuscript and for providing helpful comments, remarks and suggestions. You can find below our responses in red after each individual comment:

Due to the reordering of Sect. 3 (Results) and adding new figures, all references to text and figures of the manuscript that are mentioned below refer to the revised version, while in square brackets [] they refer to the preprint.

e.g., Figure 6 [8] refers to Figure 6 of the revised version and to Figure 8 of the preprint.

The paper "Retrieval of tropospheric aerosol, NO2 and HCHO vertical profiles from MAX-DOAS observations over Thessaloniki, Greece” by Dimitris Karagkiozidis et al., presents results of 2 MAXDOAS profiling retrievals (MMF and MAPA) for 1 year of observation (May 2020 to May 2021) in Thessaloniki. The 2 approaches are presented, with investigations of the impact of different filtering selections, and are compared to available ancillary measurements (AOD from Brewer and CIMEL, aerosols extinction profiles from a few lidar measurements and surface NO2 from in-situ data).

The paper is well written and easy to follow, and its scientific content fits the scope of AMT.

The title is however a bit misleading: we expect to learn about profiles in Thessaloniki, but NO2 and HCHO profiles are never shown in any of the figures! The paper is more about a comparison of the 2 approaches, mostly focusing on VCD and surface concentration, and comparisons to external data, when available (which is not the case for HCHO). The outcome of the study is also a bit confusing, specifying for each case the best regression statistics, but not how to deal with these data if they want to be used. Should an average of both profiling techniques should be recommended? Should we only rely on VCD and surface concentration? Should we use only one of them (eg MMF that provides AVK), but then use the bias to MAPA to estimate a (more) realistic uncertainty? Are the profiles of the 2 algorithm within their estimated uncertainties? (uncertainties of each approach are never mentioned).

It would be good that the authors provide some suggestions in the conclusions.

I would thus recommend some revision of the title and text, with some further geophysical (instead of only statistical) investigation, as described below.

I would also suggest some reordering of Section 3. The results are now presented first for VCD (3.1), then dSCD (3.2), then surface concentration (3.3) and then AVK (3.4). It would make more sense to me to follow the retrieval order: from dSCD, to profiles and AVK, and then VCD and surface extracted from the profiles. Or focusing first on the output products (VCD and surface concentration), and then some diagnostic elements (dSCD and AVK).

The study would allow to present many geophysically results, instead of only showing coherence of 2 (both are possible, see eg. Vlemmix et al., 2015). E.g. answering the following questions:

- how are the profiles themselves (is e.g. the H75 characteristic height (see eg Vlemmix et al., 2015) of NO2 lower or higher than the HCHO and aerosols one? how is it changing over the day and the seasons?)

- how is the variability within the different measured azimuths (is there an heterogeneous situation, as shown e.g. for Athens in Gratsea et al 2016? is it stronger for NO2 than for HCHO, as we would expect?).

The general comments made by Referee #2 (e.g., misleading title, vertical profiles not shown, geophysical results not presented) are very reasonable and are similar to those made by Referee #1. Based on Referee's #1 suggestions, we considered more appropriate that the paper remains in an intercomparison/validation scope, yet including additional information. The title of the paper was revised to: "Retrieval of tropospheric aerosol, NO_2 and HCHO vertical profiles from MAX-DOAS observations over Thessaloniki, Greece: Intercomparison and validation of two inversion algorithms". We included a subsection in Sect. 3 (Results) showing the seasonal mean vertical profiles retrieved by MMF and MAPA for all species. In this subsection the vertical profiles are intercompared, the variability of each algorithm is discussed along with possible sources for each species. Section 3 was reordered according to Referee's #2 suggestions: (1) dSCDs, (2) Averaging kernels, (3) VCDs, (4) Surface concentrations, (5) Seasonal mean vertical profiles.

Also, to my point of view, the paper is missing the opportunity to make the link with the previously created datasets from this instrument. It would be nice to know how much these profiling results are coherent with approaches used in the past for the VCD estimation (Drosoglou et al., 2017 and 2018, QA4ECV dataset used in Pinardi et al., 2020; Verhoelst et al, 2021; De Smedt et al. 2021). Are results similar or very different in term of VCD? E.g., see comment for P 14, line 338, or for P. 20, line 412.

The instrument that is used in this study was installed and its operation began in May 2020. Data from this specific instrument have not been used in former studies and have not been submitted to any databases (except for the FRM_4DOAS). A direct comparison with data that have been used in the past (e.g., Pinardi et al., 2020; Verhoelst et al, 2021; De Smedt et al. 2021) is not possible since these data have been retrieved using instruments of different characteristics (e.g., tracker resolution, wavelength range, spectral resolution, SNR, FOV). However, we included an extra appendix showing the comparison of the NO_2 and HCHO VCDs that are retrieved by MMF and MAPA with the VCDs that are calculated using the geometric approximation. The VCDs that have been used in Drosoglou et al., 2017 and 2018 were retrieved using pre-calculated dAMF LUTs based on RTM simulations. The NO_2 dAMFs are calculated at a wavelength that corresponds to the smaller fitting window of NO_2 (411-455 nm), because of the limited wavelength range of the older spectrographs. In this study NO_2 is retrieved at the large visible range (425-490 nm), so an additional dAMF LUT would be required, which is currently not available.

There is also a lack of reference to literature when presenting the specific results of this study and stating some "realities". (e.g., page 24, line 485 "Since the MAX-DOAS profile retrievals in the UV are sensitive only at altitudes closer to the ground*, where the lidar system is not, the profiles for 360 nm are excluded from the analysis") - *: how can we confirm this sentence?)

Done: References to literature have been included at the proper sections of the manuscript.

It would be good to also show the coherence of the lidar comparisons (Figure 12) with the AOD from Brewer and AERONET. Is the vertically integrated extinction profile coherent with the AOD? (see comment for Figure 12)

Please see relevant reply below (comment for Figure 13 [12]).

- Figure 2: please also specify other instruments location.

Done.

- Section 2.3 (or 2.6): are the 2 retrievals treating cloud filtering in the same way? are they both starting from a reduced set of cloud filtered dSCD, or is this done within the MMF and MAPA algorithms?

No cloud filtering is applied to the data prior to the analysis of MMF and MAPA. Neither MMF nor MAPA include a direct cloud flagging system. However, in MMF the stability of the retrieval is internally checked. The aerosols retrieval is performed twice: Once using the a priori profile that is defined in the settings (Sect. 2.6) and once using different a priori (and hence different covariance matrix information). If the retrieved AODs are significantly different, the trace gas retrieval is also performed twice. If the retrieved VCDs are also significantly different the elevation scan is flagged as error. Moderately thin uniform clouds do not prevent good retrievals (Frieß et al., 2019). For very non-uniform cloud conditions, it is very likely, that the two aerosol retrievals will result in very different aerosol profiles and if they have a strong effect on the trace gas retrieval, then the retrieval is flagged as invalid. For MAPA the flagging criteria might be too strict, but it is found in other studies that the elevation sequences affected by clouds are correctly flagged as invalid (Beirle et al., 2019; Frieß et al., 2019; Tirpitz et al., 2021) since some flags that are included in MAPA are sensitive to clouds.

- page 10, line 212: "the progress of the convergence is faster when using an a priori VCD or AOD below the true value" - why is this?

The reason has not been yet identified. This is found empirically, by testing the convergence behavior.

- Section 2.7: specify the location of the ancillary data - how far are they from the MAXDOAS? and mention the impact of the different fields of views.

A short introduction is included now in Sect. 2.7. Except for the in situ, all other instruments that are used in this study (MAX-DOAS, CIMEL, Brewer and lidar) are collocated on the rooftop of the Physics Department of the Aristotle University of Thessaloniki (40.634^o N, 22.956^o E), about 60 m above sea level. The location of the in situ measurement site is at a distance of ~ 1.2 km away from the MAX-DOAS and it now included in Figure 2. Differences in the retrieved products among the instruments are mainly due the viewing geometries and the retrieval technique that each instrument utilizes rather than the field of view (P.12, L251 - 252). The CIMEL and the Brewer use direct sun measurements for the calculation of the AOD, while the MAX-DOAS uses the O_4 dSCDs at different elevation angles as a proxy for the retrieval of the aerosol extinction (P.27, L538 - 540 [P22, L450 - L452]). The instruments also probe different air masses, e.g., the lidar measures only at the zenith, while the MAX-DOAS retrieves the vertical profiles by scanning at multiple elevation angles for a certain azimuth direction (P.27, L562 - 565 [P24, L473 - L475]).

- P. 12, line 259: just to have an idea, how many lidar measurements this schedule would represent in the interested time period (May 2020 to May 2021)?

The lidar measurements in Thessaloniki follow mainly the EARLINET schedule for climatological measurements with additional measurements for special events and satellite overpasses, resulting in more than 100 days of data per year. Generally, lidar measurements are only restricted by unfavorable weather conditions (rain, low clouds) and recently by system upgrade. For example, 111 days of measurements are available for the period 2019 - 2020, whilst only 35 measurements were performed between May 2020 to September 2020, when the system was set out of order for upgrading.

- Section 3: the results are presented separately for the different viewing azimuths (with no clear major difference or explation of difference between MMF and MAPA relative to the azimuth), while in Sect. 4, where the results are "validated", this information is now missing. What is used here? only one of the azimuths or an average of both or a mix of them, depending on the time of the day?

Please see the relevant replies on the comments for Figure 13 [12] and Figure 14 [13] below.

- P. 13, line 305: "the elevation sequences, for which the retrieved AOD from the MAX-DOAS inversion algorithms is greater than 1.5 are filtered-out, since such high aerosol loads are unrealistic for Thessaloniki" - is this a big proportion of data? can this be impacted by clouds, or have these been filtered before?

The elevation sequences that are passed to MMF and MAPA have not been filtered for clouds prior to the analysis. Indeed, retrievals of high AOD can be strongly impacted by clouds. The flagging that is applied to the data has proven to successfully reject retrievals under such conditions (e.g., Beirle et al., 2019; Frieß et al., 2019; Tirpitz et al., 2021). Filtering scans that result in AODs greater than 1.5 further assures that unrealistic profiles will be eliminated.

- P. 13, line 307: "Negative columns can occur in the trace gas retrievals of MAPA within the Monte Carlo ensemble and they are intentionally not removed" - add "at first/by default/..." - is this 8.5% of negative HCHO VCD points already included in the 18% valid MAPA flagged data of Table 3, or to be additionally removed ?

The fractions of 8.5% and 18% are not directly comparable. The fraction of 8.5% corresponds to the fraction of the valid-flagged data that contain negative columns, while 18% refers to the fraction of the total dataset that is flagged as valid. Since negative columns are removed from the initial dataset, the fraction of 18% refers to the data that are flagged as valid and do not contain negative concentrations.

- Table 3: add a third column with the remaining valid data percentage when both algorithms have coincident valid flags (filter #4, used as default in most of this section, if I understood well).

Done.

- P. 13, line 317: "an elevation sequence is considered valid as long as it is flagged as valid by both MMF and MAPA. This is the default flagging scheme for NO2, HCHO and AOD at 477 nm" --> this would mean flagging scheme #4 of Table 4, right?

Yes, this is true. Scheme #4 of Table 4 accounts for the data that are flagged as valid by both MMF and MAPA. The text has been revised to clarify this.

- P. 14, line 324: you mention the Orthogonal Distance Regression (ODR or bivariate least-squares) instead of an Ordinary Linear Regression (OLR or standard least-squares), but in figures 5, 7, 11 you mention linear regression. Please adapt with the correct regression type.

Done.

- P 14, line 338 "This is the first time during the Phaethon's operation that the whole elevation sequence is being used in order to derive the tropospheric VCDs more accurately": comment coherence of VCD results obtained here with respect to approaches used in past datasets (see comment above).

An extra appendix in included, showing the comparison of the VCDs that are retrieved by the inversion algorithms (MMF and MAPA) with the VCDs that are obtained using the geometric approximation (a technique that was used in previous datasets. Please see also relevant reply above (2^{nd} half of page 2).

- Figure 5: it is difficult to understand from this figure if the larger variability of MAPA results (eg for HCHO and aerosols UV) is related to the different azimuths - is MMF seeing less well the variability among the different azimuths, is MMF too sensitive or is this a false impression? are the SCD showing some systematic (?) larger signal over the city or the sea? or is this just coming from the larger variability in aerosols in the UV ? (if latter explanation is relevant, also add it to P. 16, lines 355- 356).

In the UV, the MAX-DOAS loses its sensitivity at higher altitudes faster than in the VIS. The sensitivity of the MAX-DOAS decreases with altitude and it is very limited at altitudes above 2.5 km for the species measured in the VIS spectral range or even lower (1.5 km) for the species in the UV (Figure 6 [8]). For NO_2, this is generally not a problem since the total column is dominated by the concentration in the lower layers of the troposphere (see also Figure 9 and discussion 3.5 of the revised version of the manuscript). However, HCHO can be vertically extended at higher altitudes, where the sensitivity of the MAX-DOAS is low. In the case of HCHO, OEM algorithms (such as MMF) are more prone to result in the a priori profile, while parameterized algorithms (such as MAPA) become more unstable (Frieß et al., 2019). Thus, the vertical profiles of MAPA are expected to have greater variability.

- P. 19, line 396: consider "Figure 8 shows a typical example of the calculated AVKs for each of the retrieved species. The DoF of this example retrieval are shown for each species." --> "Figure 8 shows a typical example of the calculated AVKs for each of the retrieved species, including their corresponding DoF."

Done.

- P. 19, line 399: "The averaging kernels verify that" - change "verify" to "illustrate" or something similar.

Done

- P. 20, line 412: no other source of independent HCHO is present, but this section could also be a good place to compare results from the 2 profiling algorithms to results of past VCD retrieval methods (see comment above)

Please see relevant reply above (2^{nd} half of page 2).

- Figure 9: what flagging choice is used to make this figure? from this figure, the feeling is that MAPA has systematically lower AOD @477 than the other datasets (a lot of points close to zero), which is not the case for AOD @360. I would say that the comparisons in the UV are better than in the visible...

We would like to thank the reviewer for pointing this, because there is an inconsistency in the time series of aerosols (UV) between Figure 10 [9] and Figure 7 [5]. The time series in Figure 10 [9] were supposed to use data flagged with the default flagging schemes, as described in Sect. 3 (i.e., scheme #2 of Table 4 for aerosols UV and #4 for aerosols VIS), yet, accidentally, scheme #4 was used for both species. The time series of aerosols UV is now corrected and is consistent with the rest of the manuscript. Figure 10 [9] is included to depict the different time periods that the three systems (MAX-DOAS, CIMEL and Brewer) cover during this ~1 year study. A more detailed comparison (not just visual) of the AODs between the MAX-DOAS and CIMEL/Brewer, including all flagging schemes of Table 4, is presented in Sect. 4.1 and Sect. 4.2.

- P. 21, line 435: "Compared to the CIMEL, MAPA seems to perform slightly better than MMF when its own flagging algorithm is applied to the data, with correlation coefficients of 0.70 and 0.50, respectively." - suggestion to replace by "when each algorithm consider is own flagging, with correlation coefficients of 0.70 and 0.50, respectively (MAPA for case #2 and MMF for case #1)." for more clarity!

Done.

- P. 21, line 447: "The AOD derived from the MAX-DOAS, both in the UV and the VIS range, is, generally, underestimated compared to the AOD measured by the CIMEL and the Brewer" --> add references to other studies showing that! also in P. 22, line 454.

Done.

- P. 24, line 476: "Thus, differences in the retrieved extinction profiles are expected, especially at locations with large horizontal inhomogeneities of aerosols" --> is this the case here? having a geophysically analysis (diurnal and seasonal) of the results for the different azimuths would help answer to this question. What azimuth is shown in Figure 12 for MAXDOAS?

The MAX-DOAS aerosol extinction profiles in Figure 13 [12] correspond to the elevation scans which are flagged as valid by both MMF and MAPA (scheme #4 of Table 4) that are closest in time to the lidar measurements. For the cases of 04 and 05 June 2020, the MAX-DOAS system was not scanning across all viewing directions (P.18, L418 - 420 [P. 16, L347 – 349]), so only 220^{o} azimuth is available. For the other two cases (i.e., 21-Jul-2020 and 28-Aug-2020), the elevation scans correspond to 185 and 220^{o} azimuths, respectively. The azimuth viewing direction of the MAX-DOAS are now included in the title of each panel.

- Figure 12: it would be nice to also have a comparison of the integrated aerosols profiles, to compare the lidar AOD to the MAXDOAS ones and to Brewer and AERONET (if available) for those cases.

AOD measurements from the CIMEL and/or the Brewer are not available for all cases shown in Figure 13 [12] for reasons that are discussed in Sect. 4 (see also Figure 10 [9]). However, the consistency of lidar and CIMEL AOD measurements over Thessaloniki was analyzed in the study of Siomos et al., 2018 using fourteen years of data. Periodical systematic biases (e.g., lidar overlap effect) that could affect the annual cycles, non-periodical biases that could interfere with the long-term trends and possible effects of the different sampling rate between the lidar and the sunphotometer were discussed and analyzed. The analysis resulted in consistent statistically significant and decreasing trends of aerosol optical depth (AOD) at 355 nm of −23.2 and −22.3 % per decade for the lidar (integrated extinction coefficients) and the sunphotometer datasets, respectively (Siomos et al., 2018). The AODs at 355nm measured by the lidar have also been compared with the Brewer's retrievals, showing a generally good correlation of 0.7 (Voudouri et al., 2017).

- P. 26, line 510: what MAXDOAS dataset is shown in Figure 13? all the azimuth angles together?

Since the in situ site is not located in the MAX-DOAS line of sight, hourly mean NO_2 surface concentrations from all available azimuth directions are calculated in order to avoid effects of possible horizontal inhomogeneities of NO_2.

- Figure 13: how is the fact that the MAXDOAS is situated at an height of 80m is taken into account here?

The MAX-DOAS system is located at an altitude of 60 m above sea level (not 80 m as was wrongly stated), while the in situ monitoring station is located at 174 m above sea level. The NO_2 "surface concentrations" reported by the MAX-DOAS refer to the average concentration retrieved for the lowermost 200 m layer above the MAX-DOAS location, so the in situ sampling is well within the first MAX-DOAS layer.

- P. 26, line 517: Zieger et al 2011 reference is for aerosols comparisons, it should appear in Sect. 4.1 instead of 4.2

Done.

- Figure A1: why none of the statistics for NO2 and HCHO correspond to those of Figure 7 black values? I would assume to find the same values in "O4 SF var"?!

The reviewer correctly realized that the statistics of the O_4 SF var in Figure A1 should match with the corresponding statistics of the VCDs/AODs (when no discrimination of the different azimuth viewing directions is made) that were presented in Sect. 3 (Results). However, Figure A1 presents the effect of the O_4 SF on the integrated columns (i.e., VCDs and AODs), not on the surface concentrations. Hence, the statistics of the O_4 SF var in Figure A1 should match with those of Figure 7 [5] (black values) and not with Figure 8 [7].

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