RESPONSES TO REFEREE 2

First of all, the authors acknowledge referee 2 and the editor for the time spent to review this manuscript and also for their constructive comments. The modifications are indicated by red bold fonts in the revised manuscript.

REFEREE 2

The major issue I have with this manuscript is that I don't know what is the key research question and finding of this study.

The first part of the manuscript (until page 16) presents measurements of the wildfire plume from 7 different instruments. Stratosphere, upper troposphere and lower stratosphere (UTLS), and troposphere are included in the data (Figs. 1-6), but it is not clear if there is a focus on a specific altitude range. The results are presented together with comparisons to literature, which make it difficult to distinguish between potentially new findings in this study and already existing knowledge from previous studies (Kablick et al., 2020; Khaykin et al., 2020, Kloss et al., 2021).

The uncertainty in investigated altitude range becomes evident, in the comparison of the CO observations between 9-30 km in this study with Khaykin et al. (2020) analysing stratospheric CO at altitudes above the 380 K isentrope (p14 ll11-20). 380 K are certainly stratospheric, but as Fig. 2b in this study shows, 9 km is clearly tropospheric and CO has its highest concentration in the troposphere. So, about 90 % of the CO signal shown in Fig. 4b comes from the troposphere, especially at lower latitudes. This comparison is not meaningful. It would be better to either use the same altitude criterion or calculate the column above the (lapse rate) tropopause for each data point individually.

The second part of this manuscript (pages 16 to 22) employs backward trajectories to study the origin of the air masses in the column above Reunion. Unfortunately, it is not fully clear where and when the backward trajectories were started. However, the results (Figs. 7 & 9) clearly indicate that the air masses at different altitudes (stratosphere and upper troposphere) come from different directions. But this altitude dependency is not explicitly stated in the conclusions. For the stratospheric part (~400 K isentrope) the transport pathway was investigated, but for the tropospheric part no such analysis was performed.

The discussion section (Section 5) does not contain a discussion comparing the results of this study with literature, but it rather presents an analysis of the tropospheric part of the

enhanced extinction over Reunion without explicitly stating it. Also, the altitude confusion becomes evident again. The tropopause over Reunion is at about 17 km, the lidar extinction is shown between 15-25 km (Fig. 6), but the analysed CO partial column goes down to 9 km (Figs 10, 12) and the stratospheric optical depth is shown for 15-30 km (Figs. 5, 13). There is no consistency between the data sets, which makes comparisons meaningless.

Moreover, the manuscript is hard to read and assess, as it is very long, wordy, repetitive, and does not follow ACP manuscript preparation guidelines. It contains one page long paragraphs (e.g. p3-4, p19-21), many small errors, and a large amount of references is missing in the reference list.

In my opinion, this study contains a lot of material that deserves publishing. But, I strongly recommend revising the manuscript. I'd suggest to focus on one scientific question and write down the interpretation of the material, instead of just presenting the material in a descriptive manner and leaving the interpretation to the readers. The focus could be on the observed biomass burning plume, aerosol and CO, over Reunion that has most likely two origins that are altitude dependent.

Authors: We can understand that some points were confused in the initial manuscript. The main research objective of the paper is to highlight that the Australian bushfire 2019-20 event has induced significant perturbation on the stratospheric composition over the tropical/subtropical latitudes. The previous works have mainly pointed out the transport of the Australian biomass burning (BB) plume over the high latitudes. Our paper is among the first which treats the perturbation induced by the Australian BB plume over the tropical/subtropical latitudes by focusing mainly on ground-based measurements. Through the use of numerical model (MIMOSA, FLEXPART), we clearly demonstrated the transport of the Australian BB plume over the Southwest Indian Ocean (SWIO) basin. The analysis of the numerical simulations has clearly revealed a significant increase of the stratospheric aerosol content over the SWIO basin. This result can be considered as a new finding.

The confusion on the specific altitude range of our study comes from the fact that the co-injection of CO and absorbent aerosols ends up de-correlated in space and altitude given their different properties. This is illustrated in Figure 2 with the lidar and FTIR observations at Lauder. These ground-based observations give a complementary description of the formation the BB plume to this reported in previous studies mainly based on satellites observations (e.g., Kablick et al., 2020; Khaykin et al., 2020; Ohneiser et al., 2020)[1]. Our paper brings new information on the specific altitude range associated to the injection of CO and absorbent aerosols. Then, the partial columns are calculated by considering the specific altitude range associated to CO and absorbent aerosols. We should take into account these specific altitude ranges in order to follow their spatio-temporal dispersion in the Southern Hemisphere.

We agree with the referee 2 concerning the repetitive aspect of the manuscript. As a consequence, some sections of the manuscript were rewritten. In the revised manuscript, the points mentioned here above are highlighted in order to give a clearer vision of the main research objective of this paper.

REFEREE 2

p1-l5: Please make sure to have the correct and current affiliations of all Co-authors.

Authors: The current affiliations of all co-authors were checked and corrected in the revised manuscript.

REFEREE 2

Please add the required author contributions section.

Authors: This section was included in the revised manuscript.

REFEREE 2

Please introduce all abbreviations first, e.g p2-l15: "BB" -> biomass burning (BB), p11-l11: GFAS p19-l26: "RMSC" (Regional Specialized Meteorological Center) -> Regional Specialized Meteorological Center (RSMC) p22l17: BC and OC

Authors: As suggested by referee 2, it was corrected in the revised manuscript

REFEREE 2

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[1] Kablick III, G. P., Allen, D. R., Fromm, M. D. and Nedoluha, G. E: Australian pyroCb smoke generates synoptic-scale stratospheric anticyclones, Geophys. Res. Let., 47(13), e2020GL088101, 2020.

Khaykin, S., Legras, B., Bucci, S., Sellitto, P., Isaksen, L., Tence, F. and Godin-Beekmann, S: The 2019/20 Australian wildfires generated a persistent smoke-charged vortex rising up to 35 km altitude. Communications Earth & Environment, 1(1), 1-12, 2020.

Ohneiser, K., Ansmann, A., Baars, H., Seifert, P., Barja, B., Jimenez, C and Wandinger, U.: Smoke of extreme Australian bushfires observed in the stratosphere over Punta Arenas, Chile, in January 2020: optical thickness, lidar ratios, and depolarization ratios at 355 and 532 nm. Atmospheric Chemistry and Physics, 20(13), 8003-8015, 2020

Please revise the wording of “aerosols smoke layers” (p2-l18). I'd suggest to use “smoke” or “biomass burning aerosol” and stick to it throughout the manuscript. “aerosols smoke layers” does not make sense, as smoke is a carbon containing aerosol. Please also check the use of the term “aerosols”. It is often wrong, i.e. plural instead of singular.

Authors: We understand the point of view of referee 2. Therefore, the wording of “aerosols smoke layers” was removed in the revised manuscript.

REFEREE 2

Many references are missing in the reference list, e.g. p2 l29 (Bencherif et al., 2020; Garstang et al., 1996; Holanda et al., 2020), l31 (Andreae and Merlet, 2001; Duflot et al., 2010, l33 Dowdy and Pepler, 2018, p9 l22 De Mazière et al., 2017

Authors: Referee 2 is right. The missing references were included in the revised manuscript.

REFEREE 2

There are no references to the data sets introduced in Section 2. Please provide references for the data sets including a DOI in the reference list, instead of a link to the data in the text. E.g. for CALIPSO data a DOI is available 10.5067/CALIOP/CALIPSO/CAL_LID_L1-Standard-V4-51 (https://asdc.larc.nasa.gov/project/CALIPSO/CAL_LID_L1-Standard-V4-51_V4-51).

Authors: The doi was included in the revised manuscript when it was available.

REFEREE 2

It is common standard to use past tense when describing what was done in this study, e.g. p9 l10 “... are downloaded from the NDACC website ...” -> were downloaded

Authors: It was corrected in the revised manuscript.

REFEREE 2

p16-l17: are -> were

Authors: It was corrected in the revised manuscript

REFEREE 2

p16-l18: is -> was

Authors: It was corrected in the revised manuscript.

REFEREE 2

p9-ll5-20: Are the Lauder and Reunion lidars the same? Then one description for both instruments would be sufficient. Are the retrievals different for two sites in the same network, or are they the same? If they differ, please explain why and how both data sets are comparable then.

Authors: Aerosol lidars often share common parts and similarities. For example, they often operate at the wavelengths of simple, double and triple Nd:YAG laser frequencies, namely 1064, 532, and 355 nm. Hence, the Lauder lidar and one of the Reunion lidars retrievals are comparable beacuse they provide aerosol extinction at 532 nm. However, these systems are not the same, they have different receiving optics, different electronic processing chains and even slightly different algorithms to obtain the extinction. References are provided for the reader to find details on the actual lidar designs. This is the result of different choices given that these systems are operated by two different groups and organizations.

Ultimately, the extinction profiles at 532nm can be compared to understand the evolution of the optical aerosol properties throughout their transport between Lauder and Reunion.

A sentence has been added in the manuscript to clarify this point, it reads:

"Although the lidar systems used in this study are different builds, extinction profiles at 532 nm from Lauder and Reunion can be compared to infer the evolution of the plume optical properties."

REFEREE 2

p10 ll26-32: The model and model setup descriptions are misleading and incorrect. Backward trajectories certainly do not consider dry and wet deposition as well as convection parameterization. Please describe only the processes that were used for the backward simulation. Also, the description states that backward trajectories were started over Reunion between 15 and 19 km every 0.5 km, but in the manuscript only the results for 18 and 16 km are shown and discussed. Did you release the trajectories at one latitude/longitude combination, if yes which, or did you distribute the 20000 particles over a certain area? Did you release the particles all at the same height level every 0.5 km, or did you distribute them around the height levels? Did the trajectories stop if they hit the ground, or did they bounce back?

Authors: We thank referee 2 for this comment. We can understand that our description of the FLEXPART simulations lack precision to some extent. To eliminate any confusion, we address each of the reviewer's points below:

"Backward trajectories certainly do not consider dry and wet deposition as well as convection parameterization. Please describe only the processes that were used for the backward simulation.":

The model allows to switch the convection parameterization "on" or "off" (L_CONVECTION = 1 in the "COMMAND" file). The convection is parametrized by the scheme of Emanuel and Zivkovic-Rothman (1999)[2]. This scheme is generally used in several models. This scheme is usually used in several model. The scheme uses the temperature and humidity from the wind field of ERA5 (ECMWF) to calculate the particle redistribution.

Although dry and wet deposition were already taken into account in previous versions of FLEXPART, the new version v10 saw a clear improvement in the model's processes in particular for aerosols. Pisso et al (2019) devotees in their paper a large section to describing the dry and wet deposition processes for particles and gases calculate by FLEXPART. In FLEXPART, wet deposition in term of below and in-cloud scavenging is calculated by using clouds and precipitation from ERA5 (ECMWF) and scavenging coefficient (using particles diameter, rain and snow coefficient, parameters from Pisso et al. (2019)). The dry deposition is calculated in terms of deposition velocity by taking into account the diameter and the density of particles.

In the present study, the convection parameterization has been activated. Furthermore, the removal processes by dry and wet deposition for particles have been taken into account. We note that the removal process by OH reaction has also been taken into account for CO.

"Also, the description states that backward trajectories were started over Reunion between 15 and 19 km every 0.5 km, but in the manuscript only the results for 18 and 16 km are shown and discussed."

These altitudes were chosen because the processes observed at Reunion occurred at these altitudes.

"Did you release the trajectories at one latitude/longitude combination, if yes which, or did you distribute the 20000 particles over a certain area? Did you release the particles all at the same height level every 0.5 km, or did you distribute them around the height levels."

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[2] Živković-Rothman, M.: Development and evaluation of a convection scheme for use in climate models. Journal of the Atmospheric Sciences, 56(11), 1766-1782, 1999

The 20000 particles are released from a cube with latitude, longitude and altitude coordinates (see figure below). Thus, there are 8 cubes differentiated only by their altitude coordinate with a total of 160 000 (8 x 20,000) particles released.

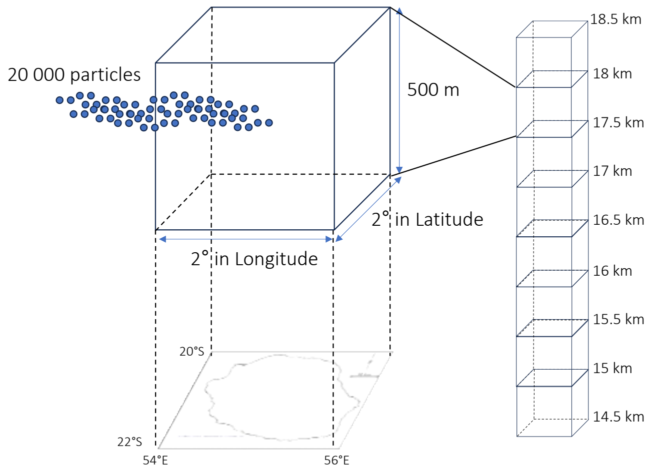

Figure 1: Illustration of the release of particles in FLEXPART

"Did the trajectories stop if they hit the ground, or did they bounce back ?"

A specific turbulence scheme is used at the boundary layer level, taking into account the vertical velocity distribution and vertical gradients in air density. Nevertheless, no information is given on the fate of particles reaching the ground.

REFEREE 2

p11 l31: Why did you drive the MIMOSA model with ECMWF meteorological analyses (which?) and not ERA5 as used with FLEXPART? Using the same meteorological input data would ensure consistency between different model simulations.

Authors: The ECMWF meteorological product used was not clearly indicated in the previous manuscript. In the present study, we used ERA5 reanalysis. This point has been clarified in the revised manuscript. Furthermore, we note that FLEXPART is also driven by ERA reanalysis. Furthermore, we note that ECMWF is an international platform where different meteorological products (e.g, ERA5, operational, INTERIM) are available.

REFEREE 2

p12 l8-p13l2: Please split the nearly one page long paragraph.

Authors: This part of the manuscript was re-written.

REFEREE 2

p13 l1-2, Fig1, Fig2: The heights of the plume observed by CALIOP (15-18 km) and by the Lauder lidar and FTIR (5-12 km) do not agree. Please comment on that.

Authors: The CALIOP track displayed in Fig.1b is dated from Jan. 1st 2020. It shows the outgoing plume originating from the powerful fire outbreak of New Year's Eve. Because these aerosols are absorbent, it has been shown that they have this interesting self-lofting capability (radiative heating). The FTIR time-series displayed in Fig.2b, also shows the overshooting feature of the CO plume as a pathway to enter the stratosphere around that specific date. Although the maximum concentration is contained in the free troposphere, enhancements are visible just before Jan.1st up to 19 km. Eventually, the co-injection of CO and absorbent aerosols will de-correlate in space given their different properties.

In addition, the FTIR and the lidar located at Lauder are pictured on the edge of the plume (Fig.1a), and they unfortunately did not record profiles on Jan. 1st. In conclusion, considering the extremely varying atmospheric context at this stage of the transport event, it is difficult to directly compare these different observations (Fig.1 vs Fig.2).

REFEREE 2

p13 l14: Why did you set the tropopause to 12 km? Figure 2 shows that there is variability in the tropopause height of ±1 km. For both the sAOD and the stratospheric CO column, this makes a significant difference.

Authors: We understand the point of view of referee 2. The characteristic of the tropopause was retrieved from available radiosonde observations (on average one radiosonde per week) by the use of the thermal definition which is based on the lapse rate (LRT) criterion. On average, the tropopause at Lauder is located at 12 ± 0.5 km during this period. We note that this value is in agreement with previous works (Bodecker et al., 1998; Griffith et al., 1998; Sakai et al., 2016)[3].

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[3] Bodeker, G.E.; Boyd, I.S.; Matthews, W.A. Trends and variability in vertical ozone and temperature profiles measured by ozone sondes at Lauder, New Zealand: 1986–1996. J. Geophys. Res. 1998, 103, 28661–28681, 1998

Griffith, D.W.T.; Jones, N.B.; Matthews, W.A. Interhemispheric ratio and annual cycle of carbonyl sulfide (OCS) total column from ground-based solar FTIR spectra. J. Geophys. Res. Space Phys. 1998, 103, 8447–8454

Sakai, T.; Uchino, O.; Nagai, T.; Liley, B.; Morino, I.; Fujimoto, T. Long-term variation of stratospheric aerosols observed with lidars over Tsukuba, Japan, from 1982 and Lauder, New Zealand, from 1992 to 2015. J. Geophys. Res. Atmos. 2016, 121, 10283–10293.

The monthly evolution of sAOD and stratospheric CO column are calculated from daily observations from lidar and FTIR, respectively. Given the sampling of the radiosonde observations (one per week), we decided to use the mean height of the tropopause for the calculation of the daily partial column (and then the monthly partial column).

REFEREE 2

p13 ll28-31, Fig. 4: Both are not comparable, because the columns range from 16 to 30 km for the aerosol and 9-30 km for the CO. CO is mostly confined to the troposphere and decreases significantly in the stratosphere. This means that Fig. 4 shows mainly tropospheric CO and stratospheric AOD. Hence, there is no correlation to be expected in the first place and many differences can be found. The difference in altitude also explains the differences described later on p14 ll15-18.

Authors: We understand the point of view of referee 2. This paragraph was re-written in the revised manuscript. Furthermore, As requested by referee 2, the manuscript was revised in order to clarify the key research question and finding of this study. The section 3 and 4 were re-written in the revised manuscript.

REFEREE 2

p14 ll15-18: Of course, you do not see the same plume as Khaykin et al. (2020). Khaykin et al. (2020) looked at heights above the 380K isentrope (~17 km) and the CO signal in Fig. 4b is from ~9-17 km.

Authors: This confusion was clarified in the revised manuscript.

REFEREE 2

p15 l27: Please explain what the different Ångström exponents mean.

Authors: The Angstrom exponent (AE) is a parameter associated to aerosols. It provides insight in the extinction evolution particles will follow with evolving wavelength. The relation is namely:

$$\begin{cases} AE = -\dfrac{log\left(\frac{x_1}{x_2}\right)}{log\left(\frac{y_1}{y_2}\right)} \\ \dfrac{x_1}{x_2} = \left(\dfrac{y_1}{y_2}\right)^{-AE} \end{cases}$$

Where x1 and x2 are extinctions of aerosol optical depth (AOD) at the two wavelength y1 and y2.

The last relation is widely used by the aerosol community using lidar, radiometer and photometers to convert an extinction at a certain wavelength to another. From a microphysics stance, a low AE is associated with an aerosol mixture driven by the coarse mode of its size distribution (coarse/large particles) while a high AE is linked to smaller sizes (e.g. fresh smoke).

Clarification has been added to the manuscript:

“The Angström exponent is a parameter informing on the extinction behavior of the atmospheric constituent with the light spectrum. It is often used to infer some microphysical properties of aerosol and in particular some information on the particle size. In general, a small Angström exponent is synonym of a coarse mode driving the optical properties of the aerosol. Insight on the Angström exponent and aerosol size as well as its relative error with respect to extinction properties are developed in Baron, et al, 2023[4] and its attached supplementary information.”

REFEREE 2

p16 l3: What is the difference between fresh and aged smoke? Please describe what you mean and how you can tell from your data.

Authors: Fresh and aged smoke can differ for obvious chemical reasons. The initially emitted smoke plume (fresh) will evolve in time while its chemical composition will change. All these changes are often referred to ageing processes. Studies have shown that these processes tend to increase the size of the particle and shaping them more spherically with time. Knowing that, the AE is expected to decrease with time within a considered smoke plume. Alternatively, different AE found in distinct layers in the atmospheric column can tell another story: the layers do not share the same origin in time and/or space.

Here is what has been added to the manuscript:

“Indeed, Muller et al, 2007[5] showed that ageing of transported smoke translates into a decreasing of the Angström exponent.”

REFEREE 2

[4] https://doi.org/10.1029/2022GL101751

[5] GRL doi:10.1029/2006GL027936

p16-ll8-14: That is quite a lot of speculation here. You could easily check if transport occurred at the Reunion lidar site by looking at the wind speeds (e.g. in ERA5) at the different altitudes.

Authors: At this stage of the manuscript, this paragraph can appear like as speculation. This paragraph gives the assumptions which are investigated in the following section (4.2). These assumptions are investigated by the use of the FLEXPART model which the dynamic is driven by ERA5 reanalysis. As a consequence, our methodology is not far than this suggested by the referee 2.

REFEREE 2

p17-ll15-21: "Given the Australian BB aerosol are mainly located in the mid-latitudes (Fig. 4a), we can reasonably conclude that the filament reaching the SWIO basin contains aerosol from Australian BB event." and "These results demonstrate that the unusual aerosol load observed in the UT-LS above SWIO is to be linked to the Australian fires." I think both sentences are kind of redundant and this part should be rephrased to be more concise.

Authors: Referee 2 is right. This paragraph was re-written in the revised manuscript.

REFEREE 2

p18 l32-p19 l3: This is speculation. Either prove or remove. The fires are clearly north of the observed enhanced CO columns and the referenced studies (p19ll 3-5, which by the way also are not in the reference list) do not suggest latitude crossing diagonal convection.

Authors: We understand the point of view of referee 2. At this stage of the paper, this is seeming speculation. As a consequence, this paragraph was removed in the revised manuscript.

REFEREE 2

p19-ll18-22: Instead of lengthy explanation, you should add the position of the ITCZ to Fig 10.

Authors: We think that the OLR anomalies should be sufficient. The ITCZ has its widest seasonal shift in the Indian Ocean region. (Fig.2). ITCZ can be widely spread, we also often talk about a double ITCZ. Ultimately, what is making the most sense, is where were the convection hotspots and to what extent they are a capable of lifting BB plume in the UTLS.

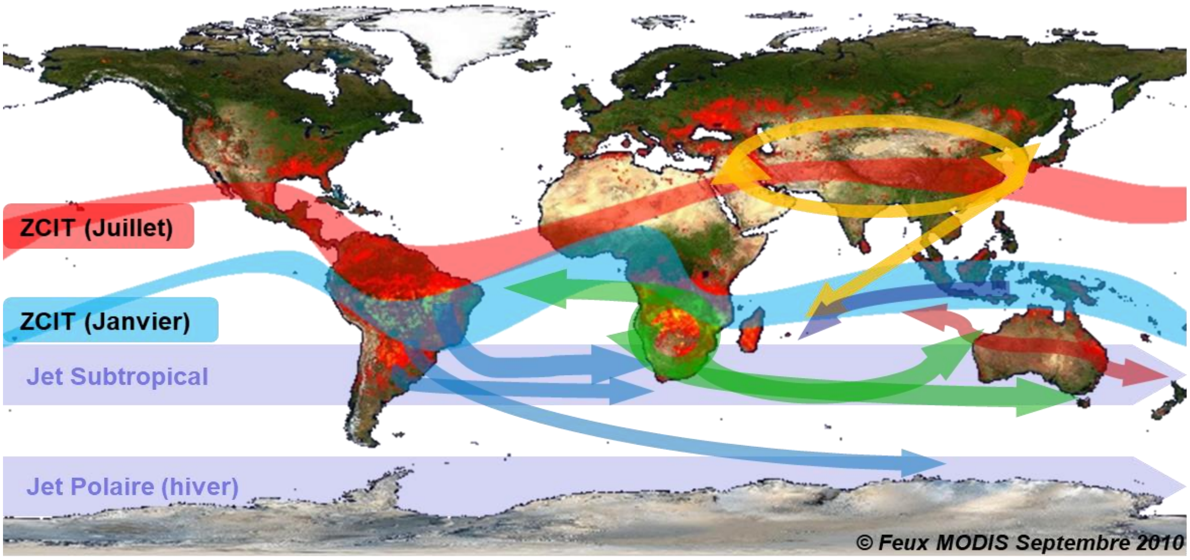

Figure 2: Illustration of the main dynamical scheme in the Southern Hemisphere

Nevertheless, we can understand the point of view of referee 2. As a consequence, the average position of the ITCZ was added to Figure 10 in the revised manuscript.

REFEREE 2

p20 l3: Which month did you use to calculate the "monthly background"? January 2020, or January 2019, or some multiannual mean?

Authors: The monthly background is calculated from available MLS observations in January between 2017 and 2019. This clarification has been included in the revised manuscript.

REFEREE 2

p20-l9-19: This is an unnecessary lengthy and incomplete description of Fig. 11 (l 14 a value is missing "15 & to %"). It would be better to present the results and implications this figure has.

Authors: It was corrected in the revised manuscript.

REFEREE 2

p33-l2: It says "FIGURES AND TABLE", but there is no table. Also the axis labels are quite small in Fig. 1b.

Authors: It was corrected in the revised manuscript.

REFEREE 2

p34: Why are there more days of measurement available in the FTIR measurements than in the lidar measurements? Was there a stricter cloud filter for the lidar data?

Authors: The light scattering by the cloud occults the scattering induced by the aerosols. It is for this reason that lidar measurements are performed in clear sky condition. This explains the fewer numbers of lidar observations in December 2019. The cloud filter allows to decrease the profiles contamination by clouds and to highlight the presence of the aerosol perturbation in the UTLS and stratosphere. Conversely, FTIR measurements are less sensitive to the cloud cover. As a consequence, there is no significant disturbance in the recording of the data.

also, Lidar and FTIR systems are completely different, and have completely different operating conditions. Direct line of sight (FTIR) vs Zenith sky (Lidar) conditions cannot assumed to be the same.

REFEREE 2

p35: Is there a typo in the y-axis label in Fig. 2a? 500 nm vs 532 nm

Authors: There is no typo in the y-axis label in Fig. 2a. The wavelength of the AOD in Fig. 2a is 500 nm. The purpose of Figure 2 is to discussion on the persistence of the anomaly on the aerosol load in the stratosphere and the atmospheric total column. It is for this reason that we did not use the Angström exponent to plot AOD and sAOD at the wavelength.

REFEREE 2

p39 and 41: The color bar labels are too small to read.

Authors: These figures were replotted in the revised manuscript.