

RESPONSES TO REFEREE 2

First of all, the authors acknowledge the referee 2 and the editor for the time spent to review this manuscript and also for their comments.

(1) Referee 2: The major issues were not revised in the manuscript. The figures shown, are rather a compilation of what is available than figures prepared for a scientific study. The partial columns are integrated over different altitude ranges 12-30 km (Fig. 3), 16-30 km (Fig. 4), 9-30 km (Figs. 4,10,12), and 15-30 km (Fig. 5,12). The aerosol AODs are presented at different wavelengths (500, 532, 745 nm) even for the same instrument, e.g. OMPS in Fig 4a and 5a, without mentioning potentially required scaling factors to make them comparable. It is impossible to compare the figures qualitatively nor quantitatively.

Authors: The revised manuscript has been strongly reorganized following the recommendations suggested by referee 2. The overall text length has been reduced by removing repetitive aspects of the manuscript. We now solely focus on one scientific question in the revised manuscript (as requested by the referee). In the present version of the manuscript, we focused on the transport of the Australian biomass burning (BB) plume over the Southwest Indian Ocean (SWIO) basin.

We investigated the transport of the Australian BB plume over the SWIO basin mainly through the use of the aerosol and CO observations over Lauder and Reunion Island. The analysis of these observations above Lauder revealed that the co-injection of CO and absorbent aerosols ends up uncorrelated in altitude given their different properties. The aerosol plume is located above the CO plume. In the stratosphere, the aerosol and CO plume are centered at 16 and 12 km, respectively. A strong ascent occurs with the BB plume due to efficient adiabatic heating. The ascension of the CO plume is limited by its photochemical oxidation which efficiency increases with altitude. It is for this reason that the dispersion of the aerosol and CO in the Southern hemisphere is investigated at different altitude range. In the revised manuscript, the dispersion of the aerosol and CO plume is investigated at 15-30 km and 9-30 km, respectively. In order to reduce the confusion, the aerosol optical properties are presented at 532 nm. The wavelength conversions to 532 nm were performed using recommended Ångström exponents. Through the use of numerical model (MIMOSA, FLEXPART), we clearly demonstrated that the Australian BB aerosol plume was transported over the SWIO basin. Furthermore, the emissions from Australia have significantly contributed on the variability of the aerosol over the SWIO basin. Conversely, the emissions from Australia have merely contributed 10% on the

variability of CO in the SWIO basin. The contribution of other sources of CO in the SWIO basin is discussed in last section of the revised manuscript.

In summary, the revised manuscript has been written with the purpose to clarify the objective of this paper and reduce the confusion on the methodology.

Referee 2: On p12 l29, the authors write that in Fig. 2a, there is a sharp increase in the extinction in the stratosphere by mid-January. But, Fig. 2, starting in December, does not show any measured profile in the stratosphere before mid-January.

Authors: We understand the point of view of the referee 2. This sentence was rewritten in the revised manuscript.

(2) Referee 2: In Fig. 2b, I can see that there are some days with enhanced tropospheric CO mixing ratios, but I cannot see the increase in the stratosphere as described on p.13 l6. To me, it is not clear why these figures are shown.

Authors: Figure 2b (Fig. 3a, in the revised manuscript) depicts the vertical distribution of the CO plume at Lauder. An enhancement of CO mixing ratio is observed in the troposphere and the lower stratosphere. Figure 0.1 shows the daily evolution of the CO mixing ratio at lower stratosphere (12-13 km) over Lauder between 1st December 2019 and 1st April 2020. Figure. 0.1 reveals a statistically significant enhancement of CO mixing ratio in the lower stratosphere from mid-December 2019. The most significant perturbation induced by the injection of CO in the stratosphere is observed in its lower part (Fig. 2b and Fig. 0.1). The CO mixing ratio value in the lower stratosphere from mid-December 2019 is on 60 ppbv which is twice time higher than background value. Above the lower stratosphere, the CO mixing ratio decrease significantly due to photochemical reactions which are more efficient with altitude (Brasseur and Solomon, 2005)¹.

¹ Brasseur, G. and Solomon, S. Aeronomy of the Middle Atmosphere: Chemistry and Physics of the Stratosphere and Mesosphere, 3rd edn, 644 (Springer-Verlag, 2005).

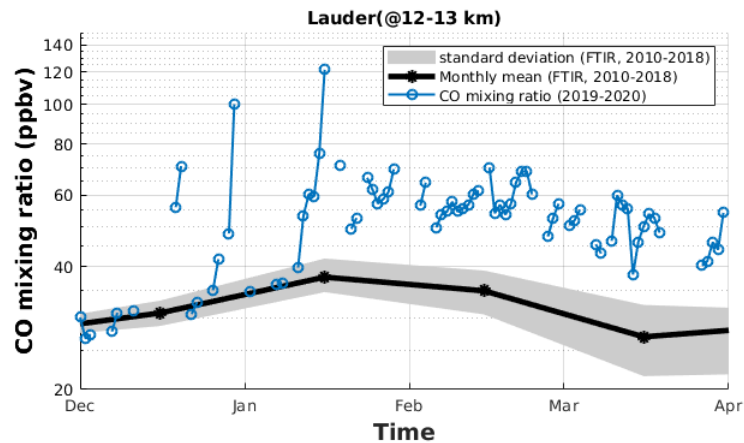


Figure 0.1: Daily evolution of the CO mixing ratio at 12-13 altitude km over Lauder derived from FTIR observations between 1st December 2019 and 1st April 2020.

Figure 2b contributes to reinforce our argument on the fact that the aerosol and the CO plume are not located at the same altitude.

(3) Referee 2: p13 l28-29: I don't understand why the aerosol and CO plumes should end up decorrelated. Fig 3 shows that both, (s)AOD and (s)CO have comparable slopes from January to March within their standard deviations. Please explain.

Authors: The aerosol and CO have been injected in the same time in the stratosphere which has induced significant perturbation on the stratosphere composition. Therefore, the evolution of the CO and aerosol in the stratospheric is correlated in time. The parameters (sAOD and sCO) pointed out by the referee 2 correspond to the amounts of aerosol and CO integrated on the stratospheric column over Lauder. It is for this reason that the sCO and s AOD are correlated in time, as it is shown in Fig.3.

Nevertheless; the aerosol and CO plume are not located at the same altitude into the stratosphere. This can be explained by the fact that the radiative and chemical properties of CO and absorbing aerosol are different. The strong absorption of solar radiation by aerosol induces a significant ascent of the aerosol plume (de Laat et al., 2012; Ohneiser et al., 2020). The aerosol plume is centered at 16.5 km due to adiabatic heating effect. Conversely, entrained CO by the pyro-convection outbreak decayed quickly to photochemical oxidation whose efficiency increase sharply with altitude (Brasseur and Solomon, 2005). The altitude of the CO plume thus ends up lower than the altitude of the absorbing aerosol layer.

Referee 2: Fig3. What is the background for CO?

Authors: The background for CO was included in the revised manuscript.

Referee 2: Fig 5: Why are no error bars shown here, as in Fig. 3?

Authors: The error bars were included in the revised manuscript

Referee 2: p10 l34: ERA5 data is not meteorological data, but reanalysis data.

Authors: It was corrected in the revised manuscript

Referee 2: p11 l4-18: I still have problems understanding the FLEXPART description. Which parameter did you use from Pisso et al. (2019)? The reference to Eckhart et al. (2017) is still missing. Please provide a reference for the turbulence and convection. Please also add reference for emission sensitivity method, e.g. Stohl et al. 2003.

Eckhardt, S., Cassiani, M., Evangeliou, N., Sollum, E., Pisso, I., and Stohl, A.: Source–receptor matrix calculation for deposited mass with the Lagrangian particle dispersion model FLEXPART v10.2 in backward mode, *Geosci. Model Dev.*, 10, 4605–4618, <https://doi.org/10.5194/gmd-10-4605-2017>, 2017.

Stohl, A., C. Forster, S. Eckhardt, N. Spichtinger, H. Huntrieser, J. Heland, H. Schlager, S. Wilhelm, F. Arnold, and O. Cooper (2003), A backward modeling study of intercontinental pollution transport using aircraft measurements, *J. Geophys. Res.*, 108, 4370, doi:10.1029/2002JD002862, D12.

Authors: Pisso et al. (2019) is a descriptive paper. The authors describe all the options included in FLEXPART and not really the chemical or physical parameters associated with lifetime of the species such as CO or BC. The model simulations involved aerosol (Black Carbon-BC and Organic Carbon-OC) and CO tracers, considering removal mechanisms such as dry and wet deposition for aerosols and OH reactions for CO. The parametrization (default values for the scavenging coefficient and the nucleation efficiency and size) for the BC was found in the paper of Grythe et al. (2017)² and the chemical parametrization for CO was in default in FLEXPART data but can be found in the reference kinetics database IUPAC (Atkinson et al., 2006³). This information is now included in the revised manuscript. We have added Eckhart et al. (2017) in the reference section.

² Grythe, H., Kristiansen, N. I., Groot Zwaaftink, C. D., Eckhardt, S., Ström, J., Tunved, P., Krejci, R., and Stohl, A.: A new aerosol wet removal scheme for the Lagrangian particle model FLEXPART v10, *Geosci. Model Dev.*, 10, 1447–1466, <https://doi.org/10.5194/gmd-10-1447-2017>, 2017

³ Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson, R. F., Hynes, R. G., Jenkin, M. E., Rossi, M. J., Troe, J., and IUPAC Subcommittee: Evaluated kinetic and photochemical data for atmospheric chemistry: Volume II – gas phase reactions of organic species, *Atmos. Chem. Phys.*, 6, 3625–4055, <https://doi.org/10.5194/acp-6-3625-2006>, 2006.

A new turbulence scheme was developed for the version 10 of FLEXPART and is well described in Pisso et al. (2017). The convective scheme described by Forster et al. (2007) was also added in the manuscript, as the Stohl et al. (2003) paper as suggested by the referee

Referee 2: Author Contributions: Who did the FLEXPART and MIMOSA simulations?

Authors: This information is included in the revised manuscript.

Referee 2: Please also indicate the software availability and a reference to the sources. I assume that with "software" in the Author Contributions section you do not mean that you wrote FLEXPART and MIMOSA?

<https://www.flexpart.eu/>

Authors: This information was included in the revised manuscript.

Referee 2: Please check the references. Many DOIs are missing.

Authors: The doi have been included in the revised manuscript when available.

Referee 2: The manuscript still contains very long paragraphs (e.g. p4 l23-p5 l31, p19 l10-20 l5)

Authors: The manuscript was re-written in order to shorten all the lengthy paragraphs.

Referee 2: many typos such as:

p5l5 Rio Grandé -> Río Grande

Please check articles throughout the manuscript, e.g. p5 l23 ...of the Australian...; ...on the variability... p5 l28 ...of a tropical... p6 l26 ...the atmosphere...

Change smoke: p3 l4; p6 l3

Please check plural s, e.g. p6 l19: altitude -> altitudes p6 l26: disturbance -> disturbances

Please use past tense when describing what you did, e.g.

p6 l27: "at Lauder is build" -> was build and many similar occurrences, especially in section 2.

p19 l5: RMSC -> RSMC please also add definition of this abbreviation.

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

p29 l13-17: remove the full stops after the last names

p29 l20-24: a new reference was put into the middle of another reference

Authors: All the typo errors mentioned by the referee 2 were corrected in the revised manuscript.