MicroCarb: Three-dimensional modelling of the $O_2(^1\Delta)$ dayglow and implications for ozone in the middle atmosphere

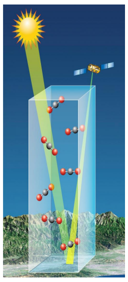
Mouhamadou Diouf[1,3], Franck Lefèvre [1], Alain Hauchecorne[1], Jean-Loup Bertaux[1], Laurent Blanot[2], Denis Jouglet[3]

[1] LATMOS, Sorbonne université, CNRS, Université Paris Saclay, Paris, France (mouhamadou.diouf@latmos.ipsl.fr)

[2] ACRI-ST, [3] CNES, Centre Spatial de Toulouse

I- The $O_2(^1\Delta)$ dayglow measured by The MicroCarb Mission

MicroCarb is a future space mission of the French national center for space studies (CNES). The launch is planned for 2024 in a sun-synchronous orbit at 650km. The objective of MicroCarb is to map, on a global scale, the sources and sinks of the greenhouse gas CO_2.



Traditionally, satellites (OCO2, SCIAMACHY, GOSAT…) measure the mean CO_2 mixing ratio (r_{CO_2}) from the CO_2 and O_2 columns:

$$r_{CO_2} = \frac{CO_2\ column}{O_2\ column\ /0{,}21}$$

Where the CO_2 column is measured in the 1.6 and 2 µm absorption band and the O_2 column in the 0.76 µm absorption band.

However, these O_2 and CO_2 absorption bands are spectrally distant. This results in significant uncertainties in the mixing ratio of CO_2 due to the varying spectral properties of the aerosols that may lead to different optical paths for photons.

The innovation in the MicroCarb mission is the addition of the O_2 absorption band centered at 1.27 µm, closer to the CO_2 bands

Problem: In this band also occurs the $O_2(^1\Delta)$ emission at 1.27 µm mainly caused by the ozone photolysis in the stratosphere and mesosphere.

The objective of the thesis is to improve the quantitative understanding and the knowledge of the $O_2(^1\Delta)$ dayglow using an advanced chemical – transport model.

II- Modelling of the $O_2(^1\Delta)$ dayglow with the REPROBUS model

REPROBUS is a Chemistry –Transport Model (Lefevre et al., 1994) with a horizontal resolution of 2°x2° that extends from the ground to 0.01 hPa, i.e. about 80 km in altitude.

- The model calculates the densities of 58 species by means of a comprehensive set of 125 gas phase reactions and 63 photodissociation rates. Heterogeneous processes are taken into account.
- The winds and temperatures used by REPROBUS are forced by ECMWF analysis.
- The chemical rate constants and absorption cross-sections are in general those recommended by the latest JPL compilation (Burkholder et al., 2019)

We implemented, in REPROBUS, all photochemical processes related to the $O_2(^1\Delta)$ dayglow as shown in fig1.

The $O_2(^1\Delta)$ is mainly produced by the photodissociation of O_3

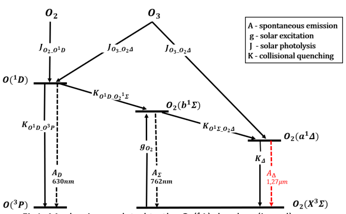


Fig1: Mechanisms related to the $O_2(^1\Delta)$ dayglow (in red) implemented in the model.

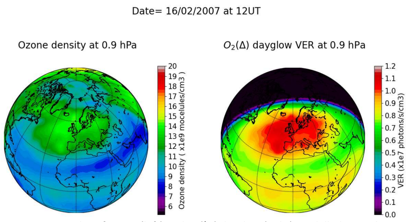


Fig2: Examples of ozone (left) and $O_2(^1\Delta)$ dayglow (right) modelled at 0.9 hPa (50 km) for 17 February 2007.

- ***The $O_2(^1\Delta)$ dayglow is strongly dependent on the ozone and solar zenith angle.***
- ***Maximum $O_2(^1\Delta)$ dayglow occurs between 45 and 50 km altitude. As the zenith angle increases, the dayglow weakens and occurs at higher altitude.***

III- Comparison to observations

III- 1 SABER instrument : Integrated $O_2(^1\Delta)$ dayglow

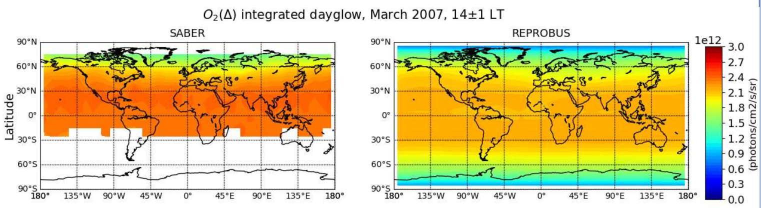


Fig3: Integrated $O_2(^1\Delta)$ dayglow between 40-80 km measured by SABER (left) in March 2007 at 14±1 LT, and calculated by the REPROBUS model (right).

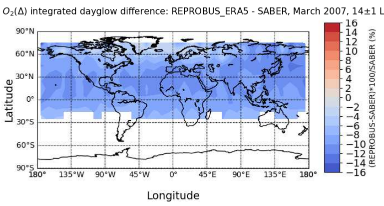


Fig4: Relative difference of integrated $O_2(^1\Delta)$ dayglow: (REPROBUS - SABER)/SABER in percent

The relative difference between SABER and REPROBUS forced by the ECMWF operational analysis, shown that the model underpredicts the $O_2(^1\Delta)$ brightness at all latitudes and seasons. On an annual average, the brightness deficit in REPROBUS compared with SABER is -11±3%. This result is consistent with the -13% deficit found by Bertaux et al (2020) who used a limited set of SCIAMACHY nadir-viewing observations as reference.

III- 2 SABER instrument : $O_2(^1\Delta)$ vertical profile

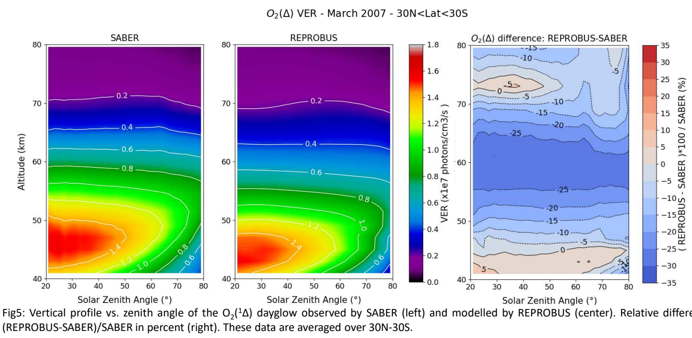


Fig5: Vertical profile vs. zenith angle of the $O_2(^1\Delta)$ dayglow observed by SABER (left) and modelled by REPROBUS (center). Relative difference (REPROBUS-SABER)/SABER in percent (right). These data are averaged over 30N-30S.

- ***The relative difference between SABER and REPROBUS shows that the model tends to overestimate the $O_2(^1\Delta)$ dayglow below the emission peak between 40 and 45 km.***
- ***Above the peak, the model underestimates the $O_2(^1\Delta)$ dayglow, with a maximum difference of -25 to -30% around 60 km.***
- ***This deficit explains the deficit in integrated $O_2(^1\Delta)$ dayglow in the model and confirms the results of Bertaux et al (2020)***

III- 3 Relationship with ozone : Comparison with MLS

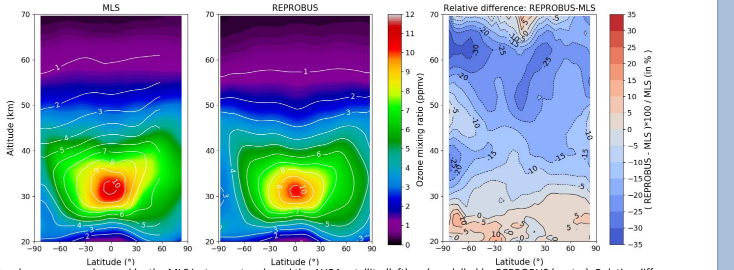


Fig6: Zonal mean ozone observed by the MLS instrument on board the AURA satellite (left) and modelled by REPROBUS (center). Relative difference (REPROBUS-SABER)/SABER in percent (right). The data shown here are for March 2007 during daytime (SZA<85°).

- ***There is an ozone deficit in the model. In the upper stratosphere the modeled O_3 is 5-15 % lower than MLS. A greater difference is found in the mesosphere, where the underprediction of O_3 in REPROBUS reaches about -30% at 60 km relative to MLS.***
- ***This ozone deficit is consistent with the $O_2(^1\Delta)$ dayglow deficit in the model. Therefore, we attribute the deficit of $O_2(^1\Delta)$ dayglow to the lack of ozone in the model.***

IV- Efforts to improve the agreement between the model and observations

IV- 1 Effect of temperature in the mesosphere

In its nominal configuration, the temperatures used by REPROBUS are forced by the ECMWF operational analyses. We investigated the effect of temperature on ozone in the mesosphere with a new simulation forced by ERA5 reanalyses (Herbach et al., 2020). ERA5 benefits from a decade of developments in model physics, core dynamics and data assimilation compared to the 2007 operational analyses.

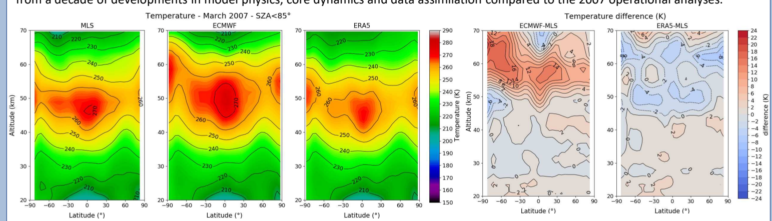


Fig7: Zonal average temperature observed by MLS (left) and analysed with operational ECMWF (center) and ERA5 (right) for March 2007 during daytime.

Fig8: Difference (K) of zonal mean temperatures between operational ECMWF 2007 (left), ERA5 (right) and MLS.

- ***There is good agreement between the ECMWF operational analysis (2007), ERA5, and MLS data up to about 45km.***
- ***However, in the lower mesophere/high stratosphere, the ECMWF operational analysis is significantly warmer than MLS with a difference of about 10-15 K. While ERA5 is in better agreement with MLS.***

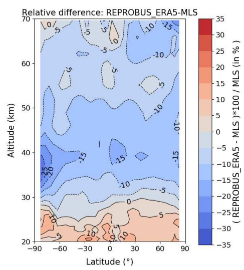


Fig9: Relative difference of zonal means of ozone modelled by REPROBUS forced with ERA5 reanalysis and measured by MLS : (REPROBUS-MLS)/MLS in percent.

- ***REPROBUS forced by the ERA reanalysis shows a considerably reduced ozone deficit. Between 55-60 km, the ozone deficit decreases from about -25% to -5%.***
- ***This increase in O_3 in the model with ERA5 is essentially due to a reduction in the efficiency of the HO_x cycles. The decrease in temperature results in a significant increase in the rate coefficient of the ozone production reaction, leading to a reduction in the abundance of atomic oxygen which is determinant for HO_x cycles.***
- ***A deficit of about 15% compared to MLS persists around 40 km altitude. This result is expected since the ERA5 temperatures do not show significant differences with the operational analysis in this altitude range.***

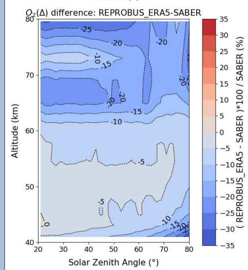


Fig10: Relative difference (REPROBUS-SABER) / SABER in percent of vertical profiles vs. zenith angle of the $O_2(^1\Delta)$ dayglow calculated by REPROBUS forced with ERA5 and the $O_2(^1\Delta)$ dayglow observed by SABER.

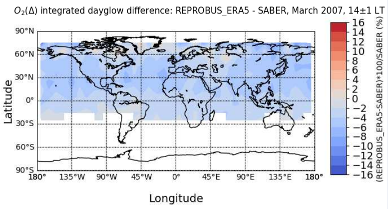


Fig11: Relative difference of integrated $O_2(^1\Delta)$ dayglow between 40-80km of REPROBUS forced by ERA5 and SABER for March 2007 at 14±1 LT. (REPROBUS - SABER) /SABER in percent.

- ***The improvement in the ozone profile with ERA5 is reflected in the $O_2(^1\Delta)$ airglow profile. The agreement with SABER is much improved, with a difference generally between +5% and -5% in the altitude range where the VER is most intense (40 -60 km).***
- ***The difference with the integrated $O_2(^1\Delta)$ dayglow measured by SABER drops from a deficit of around -11±3% with the ECMWF operational analysis to about -4±3% with ERA5, on a global and annual average.***

IV- 2 investigating the remaining O_3 deficit at 40-45 km

To try to solve the ozone deficit around 40 km, we introduced in the model a new source of ozone coming from the vibrationally excited oxygen $O_2(X^3\Sigma^-, v \geq 26)$, as theorized by Miller et al. (1994):

$$O_3 + h\nu \rightarrow \begin{cases} O_2(X^3\Sigma^-) + O(^3P) \\ O_2(X^3\Sigma^-, v \geq 26) + O(^3P) \end{cases}$$

$$O_2(X^3\Sigma^-, v \geq 26) + O_2 \rightarrow O_3 + O(^3P)$$

- ***This new source increases ozone between 35-45 km.***
- ***This allows to reduce the O_3 deficit around 40 km and to improve significantly the agreement with MLS (Fig12).***

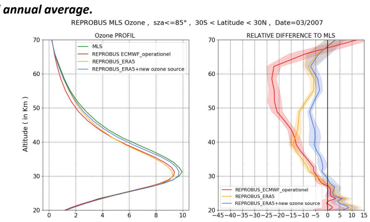


Fig12: Mean ozone profiles observed by MLS and calculated for three versions of the model (left) and their relative difference to MLS (right).

V- Conclusion

- In preparation for MicroCarb, we perfomed 3D simulations of the $O_2(^1\Delta)$ dayglow in the stratosphere/mesosphere.
- The modelled $O_2(^1\Delta)$ dayglow is significantly underestimated when the model is forced by the ECMWF operational analysis.
- This discrepancy is due to a lack of O_3(-25%) in the model between 55-65km, where we find that O_3 is very sensitive to temperature.
- The use of the ERA5 analysis, in better agreement with the observed temperatures, allows to reduce the model bias both in terms of O_3 (<7%) and $O_2(^1\Delta)$ dayglow (<4%)
- The "historical" O_3 deficit (15%) at 35-45km remains in the model, but can be mitigated by adding the proposed extra source of O_3 by vibrationally excited O_2

VI- References

Bertaux, J. L., Hauchecorne, A., Lefèvre, F., Bréon et al., (2020). AMT, 13(6), 3329-3374. https://doi.org/10.5194/amt-13-3329-2020

Hersbach, H., Bell, B., Berrisford, P., Hirahara et al., (2020). RMets, *146*(730), 1999-2049. https://doi.org/10.1002/qj.3803

Lefevre, F., Brasseur, G. P., Folkins. (1994). JGR: Atmospheres, 99(D4), 8183-8195. https://doi.org/10.1029/93JD03476

Miller, R. L., Suits, A. G., Houston, P. L., Toumi, et al., (1994). *Science*, *265*(5180), 1831-1838. DOI: 10.1126/science.265.5180.1831