

Interactive comment on “Impact of land convection on troposphere-stratosphere exchange in the tropics” by P. Ricaud et al.

P. Ricaud et al.

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Responses to Reviewers' comments

First of all, we would like to thank all the reviewers for their very helpful comments and suggestions for improving the quality of our manuscript. We have thoroughly modified the manuscript and the associated Figures for taking into account all the comments from the reviewers. Since some of them were pointed out by several reviewers, we have highlighted 10 points listed below, in addition to the modified abstract, introduction and conclusions. The Figure numbering refers to the numbers specified in the new version of the manuscript.

A. GENERAL RESPONSE

1) Pressure/altitude vs. isentropic surfaces

The geographical distributions of N₂O, CH₄, and CO in the tropical UTLS are available at three levels: i) at 19 km (66 hPa) for SMR and HALOE or 70 hPa (18.7 km) for MLS in the lower stratosphere, ii) at 17 km (94 hPa) for SMR and HALOE and 100 hPa (16.6 km) for MLS, around the tropopause and iii) at 150 hPa (14.2 km) for MLS and MOPITT CO in the UT. Note that since the tropopause height varies in longitude, the 17 km/100 hPa level is below the tropopause over the Western Pacific but above it over continents. For removing the impact of the longitudinal as well as the latitudinal change of height of potential temperature surfaces, trace gases measurements have been interpolated on the 400-K surface (approximately 1.5 km above the tropopause) for all the species, and on the 360-K surface (approximately 1.5 km below the tropopause) for MLS CO. The trace gas distributions simulated by MOCAGE at three isentropic levels, 400 K in the stratosphere, 360 K near the tropopause and 340 K in the upper troposphere for the MAM season in 2002-2004 averaged within bins of 10° latitude and 30° longitude, are shown in Fig. 9 (N₂O), Fig. 10 (CH₄) and Fig. 11 (CO). As shown in Fig. 2, the potential temperature at 17 km is the lowest over the Western Pacific but the highest over Africa and South America, meaning that the tropopause defined as the 370-K surface (Fig. 2c) is on average 0.6 km higher over the Western Pacific than over the convective African and American continents. Since an air parcel moving adiabatically along the 370-K surface will be uplifted above Western Pacific compared to land areas, the concentration at constant altitude of a tropospheric-origin trace species should be larger over the Western Pacific than over continents.

2) Top of convection

The top height, frequency and diurnal cycle of convection captured by MOCAGE have been studied by calculating probability density function (PDF) of the top of convective events. The results for three convective regions between 10°S-10°N, Western Pacific (135°E-165°E), South America (45°W-75°W), and Africa (15°E-45°E) are shown in Fig. 16. The convection frequently reaches 100-300 hPa (82%, 76% and 65% at the 3 locations, respectively), less frequently 150 hPa (26%, 20% and 12 %, respec-

tively), on rare occasions (a few %) 125 hPa, and never above. Besides the slightly smaller daily average frequency, the main difference between continental and oceanic convection resides in the diurnal cycle. A maximum development is observed around 13:00-15:00 hours (local time) in Africa and South America afterwards the altitude of cloud top drops in the afternoon and in the evening. In contrast, the Western Pacific shows a weak diurnal cycle with a maximum height of convection around 06:00-09:00 hours (local time). At the local time of maximum development, the frequency of convective systems reaching 300-150 hPa increases to 92% over the Western Pacific and 82% over both continents. In summary, the altitude reached by convective systems simulated in MOCAGE is roughly the same over the Pacific than over Equatorial Africa and South America, although the frequency of occurrence is slightly larger in the first case. Very consistent with observations, i.e. OPFs of the TRMM radar (Liu and Zipser 2005), the main difference between continental and oceanic convection is the presence of a strong diurnal cycle over land with a development peaking in the early afternoon, in opposition to a change of smaller amplitude peaking in the early morning over ocean. However, it must be emphasised that due to its relatively broad horizontal resolution ($5.8^\circ \times 5.8^\circ$), MOCAGE can hardly capture the local vertical transport associated with mesoscale convective systems, as well as small scale local overshooting in the stratosphere associated to land thunderstorms.

3) Emissions

The horizontal distribution of a trace gas lifted in the upper troposphere depends on the location of convective systems but also on the distribution of the gas at surface levels, that is, on the location and intensity of its sources and on their horizontal transport near the surface and finally on their life time. Namely, the concentration of a long-lived species such as N₂O of 130-140 year lifetime should be more homogeneous than that of CH₄ of 8-10 year lifetime, or CO of 2-month lifetime only. For better testing the impact of the inhomogeneity of surface emission on the distribution for the species in the TTL, we have also considered CFC11 a long-lived species (~50 5 year lifetime) emitted only

in the Northern Hemisphere industrialized countries, and CH₃Cl a long-lived species (~1.5 year lifetime) of both land and oceanic origin, whose distribution in MOCAGE is relaxed between 550 hPa and the surface towards a globally-averaged value specified in WMO (2002), with a relaxation time of 7 days. As an example of the highly contrasted source distribution, Fig. 12 shows the surface emissions of N₂O, CH₄, CO and CFC11 in April 2003 (see section 3.1). Though oceanic emissions slightly contribute, the main sources of N₂O are the regions of dense vegetation, Amazonia and the Congo basin in the tropics and South-East Asia. The sources of CH₄ are mainly wetlands in South-East Asia, and for a smaller part biomass burning, which explains the emissions in Africa and South America correlated with TRMM/VIRS fire counts during the same season. Note that equatorial primary forests are not sources of CH₄ in this inventory. This is in contradiction with recent ENVISAT/SCIAMACHY observations indicating significant emissions from evergreen forests in South America (Frankenberg et al., 2006) which may indicate a systematic underestimation of CH₄ also in Africa and the Indonesian Islands. CO emissions in the tropics are mainly related to biomass burning, though the largest source is pollution in populated and industrial areas. Most intense CO emissions in the inventory are those located over India, South-East Asia and China, although it must be remembered that their intensity could be highly variable from one year to another. Finally and although not measured, Fig. 12 shows the sources of CFC11. Apart from sources location and intensity, the distribution of trace gases at the surface is also controlled by horizontal transport. Figure 13 shows the average ECMWF mean wind field at 1-km height during the MAM season in 2002-2004. Equatorial convective areas are also regions of convergence of North-East and South-East oceanic trade winds on both sides of the ITCZ. However, Equatorial Africa and to a lesser degree the North-West part of South America are different, where advected air masses are passing over continentals surfaces instead of oceans and therefore enriched in continental gases compared to oceanic air masses. The influence of horizontal transport can be seen in the MOPITT CO at 850 hPa, displaying a large mixing ratio on the west side of Africa and South America extending to the adjacent ocean, in

contrast to the east side of the continents where the CO concentration is close to that of the ocean, features well captured by MOCAGE. Besides from being a source region itself, Equatorial Africa appears as a region of accumulation and thus of highest land trace gases concentration compared to other tropical continents. Figures 14 and 15 show the distributions of CFC11 and CH₃Cl as calculated by MOCAGE, respectively at 400 K, 360 K and 340 K in MAM 2002-2004. Although emitted in the northern mid-latitude only, the simulated CFC11 distribution at 400 K is very similar to that of N₂O (Fig. 9a). Furthermore, it shows a maximum over Africa at 340 K and 360 K, similar to that of N₂O, although there is no emission there, demonstrating the convergence of surface emitted species over that continent. The same could be observed on CH₃Cl showing higher concentration at 340 K around 15°S above the convergence zones of the Southern Hemisphere, drifting to 5°S at 360 K, but in that case, and in contrast to all other species, a relative maximum over the Western Pacific. However, the difference vanishes at 400 K, where the Figure is the same as for all others displaying a zonal band with two slight maximum areas over Africa and the mid-Pacific but of very limited contrast of ± 1 ppbv. At this altitude level, the signature of the source location disappears. In conclusion, although of different sources distributed differently and of different lifetimes, the tropospheric-origin trace gas distributions are very similar when reaching the tropopause level displaying all a relatively enriched area over the African sector, extending to South-East Asia for CH₄ and CO having intense sources there and to the Western Pacific for CH₃Cl largely of oceanic origin. Although the source location and intensity play a role, this suggests that the main driver for the maximum mixing ratios over convective areas at equator is the convergence associated with these at surface levels. However, the influence of sources and horizontal redistribution reduces rapidly at upper levels at 400 K in the lower stratosphere, where all species are showing almost the same feature: an equatorial belt with two maximum areas over Africa and the East Pacific, but of very little contrast. At these levels, the distribution of the species is largely the result of the uplift by the residual circulation of the meteorological analyses.

4) Flux in the UTLS

An indicator of the contribution of convection and residual vertical transport provided by ECMWF is the mass flux of a species at different levels. Figure 17 shows the N₂O vertical flux at 70 hPa, 100 hPa, 150 hPa, and 300 hPa, expressed in Mton/cell of MOCAGE (5.8°x5.8°), where positive (negative) values mean upward (downward) fluxes. At 300 hPa, areas of upward vertical transport match the location of intense convective systems and the ITCZ, and large areas of subsidence are observed elsewhere. The largest upward transport is located over the Western Pacific. Although the flux drops by a factor 3, the picture is very similar at 150 hPa, with the remarkable exception of South America, where it becomes negative over Amazonia. At 100 hPa, the flux drops again by a factor 2, though there is still some correlation between upward flux and convection. Note that the maximum upward flux is starting being displaced from the Western Pacific to the Central Pacific following the ITCZ. Finally, at 70 hPa in the lower stratosphere, the flux dramatically reduces by a factor 5 compared to 100 hPa and becomes positive on average over the whole $\pm 30^\circ$ tropical band, not showing any more correlation with convective regions. Convective transport stops at 100 hPa in MOCAGE.

5) Horizontal and vertical transports

The most intriguing feature in the MOCAGE simulations is that despite of the fact that the most intense upward flux is located over the Western Pacific in the troposphere, all species always show a maximum concentration at the tropopause over Africa, and South-East Asia for those of more intense emission there, suggesting a strong influence of horizontal transport within the troposphere. Figure 18 displays an altitude-longitude cross-section of the average concentration of CO, CH₄, N₂O and CFC11 in MAM 2002-2004 together with zonal/vertical winds shown by arrows, where the vertical component is amplified by a factor 100. Also plotted is the zonal variation of AVHRR OLR showing deepest minima over the Western Pacific and South America, and two others of a little less amplitude over Africa and Indonesia. At surface level, as discussed in section 3.3.1, the CO concentration is the largest over Africa and then South

America and Indonesia. The signature of a fast convective transport between the surface and the UT could be seen over the three areas, resulting in peak mixing ratios at 12-13 km more pronounced over Africa. Then, the CO concentration drops sharply around 17 km. As already seen in the previous section, the upward transport stops over South America at about 13 km. The larger CO in the UT over Africa is clearly related to the larger surface concentration. The CH₄ concentration at the surface is the largest over Africa and then South-East Asia but, as already discussed, smaller over South America. As for CO, fast convection results in a maximum around 12-13 km, but of larger amplitude than at the surface, suggesting some enrichment by horizontal advection from other longitudes. Note that, in the mid-to-upper troposphere, meridional winds diverge from the Equator to the tropics (Hadley cells). Furthermore, there is no indication of significant CH₄ penetration in the LS above 17 km. As for CO, the CH₄ African maximum in the UT is clearly related to its higher concentration at the surface, but requires also some enrichment by advection from other longitudes. The longer lifetime of the N₂O species makes its concentration more homogeneous in the UT and of weaker gradient at the tropopause. A maximum concentration is observed in the UT over Africa but not directly related with an increase at the surface. Africa is not an area of high N₂O surface concentration. An advection from other longitudes is required. Finally, a maximum CFC11 is also present in the UT over Africa, although the sources of CFC11 are all located at northern mid-latitudes. As for N₂O, this implies horizontal advection in the mid-troposphere before convective uplift. The slightly depleted amounts of surface N₂O and, to a lesser extent, of surface CH₄ and CFC11 over South America and Africa compared to the maximum CO, can be explained by relative intensity of the sources (~100 molecules m⁻² s⁻¹ for CO, ~1-10 molecules m⁻² s⁻¹ for CH₄, <1 molecule m⁻² s⁻¹ for N₂O) compared to the convergence of oceanic air of poorer concentration at the base of convective systems. As shown by the wind vectors in Fig. 18, the enrichment of concentration between 10-14 km (300-150 hPa) over Africa and more generally the Western hemisphere between 90°W-120°E, is largely resulting from a convergence in the upper troposphere compared to the divergent Pacific

area.

6) Rapid overshooting vs. slow radiative heating in the UTLS

The MOCAGE N₂O, CH₄ and CO profiles over Africa, South America, Indonesia and Western Pacific are shown in Fig. 20 together with the observations of the space-borne instruments over Africa in black and the Pacific in red. Because of their little consistent high and low biases at 100 hPa and 70 hPa (see section 2.3), the currently available MLS version 1.5 N₂O retrievals are ignored in this representation of vertical gradients. Although observed and modelled concentrations at the tropopause are very consistent over Africa, the most remarkable feature shown in the Figure is the large overestimation of N₂O and CH₄ over the Pacific at and above the tropopause and, surprisingly, of the vertical gradient of the species in the stratosphere, weaker in the model even over the most convective area in Africa. Moreover, and although the HALOE CH₄ average concentration in MAM over Central America is consistent with the WB57F measurements in January 2004 over Costa-Rica (Richard et al., 2006) both showing reduced concentration at 100 hPa compared to the tropospheric value, there is no indication in the model of fall-off above 14 km in the UT, at any longitude, such as reported by the aircraft. In contradiction to observations, the vertical transport in the UTLS in the model does not vary in longitude and in addition is too strong. This is consistent with the known overestimation of the vertical wind above the zero net radiative heating level at 14 km derived from the residual circulation in meteorological models (Vaughn and Hall, 2002) and in MOCAGE in particular (Teyssèdre et al., 2007) implying an overestimation of the average mass flux at the tropopause in the tropics. In contrast to this, the observed large longitudinal variation of concentration at both 17 and 19 km would imply a vertical transport of the same amplitude as in the model, but limited to convective regions. The proposal for this localised additional transport is the fast convective overshoot of tropospheric air in the lower stratosphere as observed in February 2004 over Brazil (Pommereau and Held, 2007) successfully captured by high spatial resolution Cloud Resolving Models (Chaboureaud et al., 2007; Grovesnor et al., 2007).

The stronger uplift over Africa in MAM very consistent with the maximum TRMM radar OPFs over the same region at the same season strongly supports the suggestion.

7) Vertical resolution of satellite measurements

The vertical resolution of the limb-viewing CO, CH₄ and N₂O instruments in the UTLS is ranging from 2-4 km while it is about 6-8 km for the CO nadir-viewing MOPITT providing independent information into two layers only in the troposphere. Figure 19 displays the theoretical impact of the vertical resolution on the N₂O field at 17 km provided by MOCAGE, and by convolution of the MOCAGE profiles with a triangular filter of 1-km, 2-km (similar to ODIN) and 4-km (similar to MLS) half-width. The degradation of resolution results in: i) a sharper meridional gradient consistent with observations (Fig. 3 and 4), and ii) the lessening of the average value and the smoothing of the longitudinal contrast between maximum and minimum concentrations, but no change in their location.

8) Convection into MOCAGE vs. ECMWF

Like other Chemistry and Transport Model, MOCAGE needs meteorological parameters to constrain its atmosphere for both transport and photochemical processes. For the simulations presented here, all these parameters are taken from European Centre for Medium-range Weather Forecast and correspond to operational analyses between years 2000 and 2005. They vertically extend upon 60 layers from the surface up to about 70 km of altitude. These meteorological fields are surface pressure, temperature, specific humidity and horizontal winds (zonal and meridian components). The vertical winds are then deduced by solving the continuity equation to ensure mass conservation. With these data prescribed every 6 hours, then MOCAGE calculates three-dimensional evolution of tracers, by advecting them using a semi-Lagrangian advection scheme and by diagnosing convection to take into account sub-scaled phenomena by using different criteria based upon the meteorological forcings. Chemistry is computed gathering species into families, as described in Brasseur and Solomon (1987). Two

convection schemes can be used within the MOCAGE model. Both are mass flux schemes. The first one is a simplified version of Tiedtke (1989) where downdrafts, inside-cloud subsidence, and organized entrainment over cloud base are neglected. Penetrating convection is assumed to occur when both a deep layer of conditional instability and a large-scale moisture convergence exist (therefore implying surrounding columns influences). The second scheme is the one from Betchtold et al. (2001). It is also a mass flux scheme, but the parameterization is slightly more complex than Tiedtke's as downdrafts, freezing and melting are considered. Calculations are made for a single column (i.e., no influence of neighbouring columns). Convection is activated or not by evaluating a single air parcel that is lifted from the ground to its condensation level. Contrarily to the Tiedtke scheme, scavenging (below and inside cloud, for both stratiform and convective clouds) is taken into account when using Betchtold scheme. This is of primary importance for long-term simulations of chemical species that are soluble (such as nitric acid). More details about these convection schemes used within MOCAGE, and validation results when simulating ^{222}Rn and ^{210}Pb fields, can be found in Josse et al. (2004). The present run was performed using the Betchtold scheme. Finally, note there is no parameterization of overshoot processes in the model.

9) One single season (MAM)

The use of satellite observations was part of the HIBISCUS programme dedicated to studying the impact of tropical convection on the lower stratosphere, for which ODIN observations were reinforced and long-duration balloons flown during the same period in March-April 2004.

10) Quantitative Analysis

Going a step farther from the present analysis in order to be more quantitative would require in parallel the use of two tools. Being given that MOCAGE is a tropospheric-stratospheric CTM model, we need 1) to get satellite measurements of long-lived

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species in the entire UTLS not only LS for N₂O and CH₄ (as in a present work) since CO in the UTLS is too much affected by surface emission, and 2) an assimilation tool to optimally combine measurements and model outputs i.e. constrain the model with the measurements. Microwave limb-viewing measurements of N₂O are so far restricted to pressure below 100 hPa but IR limb-viewing measurements of several long-lived species can indeed be retrieved in the UT (e.g. ENVISAT/MIPAS and ACE). This will definitely improve the study in quantitative interpretation. The MOCAGE model can be constrained by limb-viewing measurements (O₃, N₂O, etc.) using the PALM coupler developed at CERFACS (Toulouse, France) with a 3DFGAT assimilation method. Once measurement sets are debiased, it will then be possible to quantify the actual mass exchange between the UT and the LS in the tropics during the MAM period.

B. SPECIFIC RESPONSE TO REVIEWERS' COMMENTS

Response to Reviewer 1's comments:

1. Vertical resolution of the measurements. "As the authors explain, the vertical resolution of the measurements is between 2 and 4 km (and even poorer for MOPITT CO), and therefore it is not clear to which extent the enhancements shown in the measurements are stratospheric, within the TTL or even in the troposphere." Points 1) and 7).

"And even if it is STE, it still could be convective uplifting to the upper troposphere followed by slow adiabating uplifting." Points 2) and 6).

2. Horizontal and vertical inhomogeneity of trace gas distributions. "How large the anomaly is at 17 km then depends on the vertical profile of the species, and one could argue that part of the observed signal (and lack of it) is related to the distribution pattern of the species in the troposphere." Points 3), 4) and 5).

3. Model comparison. "It is not clear to me what we learn from the comparison with the model simulation. If the problem in MOCAGE is the underestimation of the maxi-

mum altitude of convective uplifting, what is the reason - a parameterization used, the ECMWF data used as input, the spatial resolution of the model? Points 1) - 10).

Minor comments. a. HALOE : " profiles are continuously retrieved " . Modified in the new version.

b. HIBISCUS : link with the HIBISCUS campaign (?) Point 9).

c. Fire counts Indeed, the fire count map is an accumulation of all fire counts over March-May 2002-2004.

Response to Reviewer 2's comments:

1. Demonstrate that "rapid uplift over land convective regions" is the dominating process of STE. Points 4) and 5).

2. Slow ascent vs. Convective overshooting. Point 6).

3. Sources Point 3).

4. Slow ascent vs. inhomogeneities vs. PT surfaces Point 1).

Other comments 5. p. 3279: "The authors should say which month the average temperature of 191 K corresponds to. There is a large seasonal cycle." As mentioned all over the text, all the study is performed considering the March-April-May 2002-2003-2004 period. It is consequently the case for the average temperature of 191 K. We have clarified this point.

6. p. 3279: Definition of "overshooting" and "STE". The definition of "TSE" and "overshooting" has been inserted in the new version of the manuscript as transport process from the troposphere to the stratosphere crossing the cold point tropopause and energetic convection overshooting the tropopause, respectively.

7. p. 3280: convection into MOCAGE vs. convection into ECMWF. (see also point 3 of reviewer 1). Additional convective transport beyond that present in the ECMWF winds?

Point 8).

8. p. 3282: Fluxes. "How high does convection inject these species in the MOCAGE model? Are there any regional variations? More quantitative approach. (See also response to point 3 of Reviewer 1.)" Point 2).

9. p. 3282: "It mentions that the land/ocean contrast for N₂O is about 15 ppbv. What instrument does this refer to? Obviously the huge differences between the measurements complicates interpretation." It refers to the Odin/SMR instrument. This is clarified in the new version.

10. p. 3272. CH₄ sinks. CH₄ lifetime is 8.9 years in the troposphere, and 120 years in the stratosphere. But vertical distribution of CH₄ lifetime (Brasseur and Solomon, 1985) shows 120 years around 20 km and less than one year at 50 km.

11. The titles of the legends were written in a small gray type. The titles of the legends were written in black. We hope that they are becoming more visible in the new version.

12. Many of the Figures use different colour schemes. Clarified in the core of the text and in the Figure caption. The new Figures for a considered species have rigorously the same colour Tables.

Response to Reviewer 3's comments:

Major problems. 1. "Method is flawed, since it depends both on the intensity of the convection and on the abundances of the considered gas in the troposphere." "They need to make the paper more quantitative, with more rigorous evaluations of the various fluxes and show that the magnitude of the convective flux is greater than the slow ascent flux." Points 1) - 10).

2. Surface variations in the source gases. Point 3).

3. Vertical resolution of satellite data. Point 7).

4. Horizontal vs. Vertical Transports. "What is the role of lateral transport of strato-

spheric air into the tropics? Descent over convection in the Western Pacific (Sherwood, 2000), this might also explain lower values of CO, CH₄ and N₂O by bringing stratospheric air down into the TTL. Does their mechanism contradict this one? Or can they coexist?" Point 5).

Less serious problem. 5. Isentropes. "Best coordinate system for this type of analysis is potential temperature." Point 1).

6. Time of year. "They are drawing general conclusions from a single time of year (late winter/early spring). What reason do we have that other times of year also show this pattern? The authors should pick another time of year when there is little biomass burning in Africa and see what the CO fields look like." Point 9).

7. and 8. Writing. "There are many places where the writing is ambiguous." The writing is clarified in the new version of the manuscript.

9. Top of the TTL. Point 2).

10. More discussion of the convective scheme: "1) which scheme is being used?, 2) does it include overshooting?, 3) if not, how high is convection going?, 4) is it getting into the TTL?" Point 8).

11. Missing references. Dessler (2002), Gettelman et al. (2002), Sherwood (2000). These references are included in the new version of the manuscript. Note that Schoeberl et al. (2006) was already mentioned in the previous version of the manuscript.

Response to Reviewer 4's comments:

"I think the paper by Ricaud et al. and the subsequent interactive comments concern an important issue that the scientific community has to deal with.""[From ECWMF data], I would expect to find also the highest long-lived trace gas concentrations exactly there [over MC/WP]. How can it be that this is clearly not the case?""Tracers with predominantly continental sources and strong land-ocean contrasts lead to a bias in favour continental convection.""[Looking at AVHRR-OLR] This shows lowest OLR over

MC/WP and South America, but not over Africa. I find this hard to reconcile with the author's hypothesis and think they need to address this issue." Points 1) - 10).

Responses to A. Tuck's comments:

We thank you for your comments and your suggestion regarding the CH₄ WB57 measurements above Costa Rica. Here is our reply. Entry of tropospheric air in the stratosphere. It is true that the N₂O and CH₄ maxima over land in the LS as shown in the paper are much more corresponding to water vapour maxima than minima as reported for by Rosenlof, JGR, 1997 from HALOE or Read et al., JGR, 2001 from MLS/UARS. But, in our mind, the question would be more that of a possible hydration of the LS, e.g. by injection ice particles, than that of a contradiction between dehydration and TSE. Indeed, as pointed out by Referee #3 (and by Richard et al., 2006), it is well recognised that dehydration is a process independent of vertical transport across the tropopause. Since we did not deal with water vapour in the paper, we prefer leaving out the issue, at least for the moment. Methane fall-off in the UT. This is a very useful comment, which we missed in the paper. We have no measurement in the UT but at 100 hPa showing significantly less CH₄ above Central America (about 1.64 ppmv) in MAM, consistent with the WB57F measurements in January, than over Africa (1.76 ppmv). The model is not capturing more than 0.01-0.02 ppmv between the two areas at 100 hPa as well the fall-off in the UT over Central America or the East Pacific or anywhere else. The very similar picture for N₂O suggests that it is not a question of source but of transport. However, if the difference was due to horizontal exchange with the mid-latitudes it should be in principle very well captured by MOCAGE forced by ECMWF known to provide excellent horizontal winds. Since it is not the case, another explanation could be an overestimation in the model of the vertical transport at all longitudes in the UT above the zero net radiative heating level around 14 km. We have added reference to the WB57 CH₄ fall-off in the UT in Section 4 and discussed the possible explanations for its absence in the model.

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