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Interactive comment on "Sensitivity of the recent methane budget to LMDz sub-grid scale physical parameterizations" *by* R. Locatelli et al.

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Dear reviewer,

We are very grateful to you for reviewing the manuscript and for submitting helpful comments and suggestions to improve the text. Here we respond point by point to your comments and questions.

The co-authors.

General comments

- **1. The manuscript refers to an unpublished manuscript of Monteil et al. However, in most cases it would be better to refer to a JGR paper that has already been published (Monteil et al, 2013).**
  Yes, we now refer to the paper of Monteil et al. (2013) according to this comment.

- **2. That paper provides a quantification of the bias using the TM5 model, which would be useful to compare with the results obtained in this study using LMDz.**
  Monteil et al. (2013) have quantified the bias between surface measurements and simulated surface concentrations using fluxes coming from GOSAT-only inversions. They have found that "Full-Physic" and "Proxy" GOSAT-only inversions lead to an overestimation of global mean surface CH_4 mixing ratios by respectively 16.9 and 6.9 ppb. However, they give little information on the latitudinal distribution of these biases, even if they seem larger in the southern hemisphere (see their Figure 5). In our work, we quantify the bias using a similar approach (comparison between optimized and measured surface concentrations). After our different inversions, we find a global mean bias of -4, +40, +38 and +41 ppb for LMDz-19, LMDz-TD, LMDz-AR4 and LMDz-KE at the surface. Consequently, the bias found when using the TM5 model (which has 25 vertical levels) in inversions is larger than in LMDz-19 inversions, but lower than in the different inversions using LMDz with 39 levels (LMDz-TD, LMDz-AR4 and LMDz-KE). Some indications may be found in Patra et al. (2011), but they refer to an old version of LMDz (19 vertical levels and old parameterizations). We investigate here the possible cause of this large increase of bias when moving from LMDz-19 to LMDz-39 and point towards the quality of tropospheric-stratospheric exchange both in the model and in the retrieval. In the revised version of the manuscript, we add

a paragraph describing the different results found with TM5 and LMDz inversions.

- **3. In the conclusions, it is mentioned that transport model errors lead to flux errors up to 50% at regional scales, but I do need see that back in any of the presented results.**
  Actually, we wrote in the "Conclusions" that uncertainties in parameterizations lead to flux errors of 5.2, 10.7 and 8.2% for respectively BG, EXT and PR-LEI inversions. Locatelli et al. (2013) used 9 transport models (with different parameterizations, resolution, advection scheme, etc.) to estimate the total error due to transport modeling in inversions. They found that spreads in regional fluxes could range from 23% to 48% of emissions depending on the regions. By taking the ratio between this study (5-10%) and the previous study (23-48%) we estimate that the error due to the vertical parameterizations in one model (LMDz) explains, on average, 24% of the total transport model errors at regional scales, and that they can reach more than 50% in some specific regions. For example, the spread due to total transport model errors in inverted fluxes for South America reaches 48% of the emissions of this region. The spread in South America due to parameterization uncertainties reaches only 9.8% in PR-LEI inversion, which means that parameterizations explain 20.4% of total transport model errors.
  We have clarified this point in the updated text.

- **4. Looking at Figure 5, I wonder how significant the differences are, given the posterior flux uncertainties and the change from the prior. The figure shows a horizontal bar, which is not explained in the caption, but may actually be the prior. It is not only relevant to assess the uncertainty in the regional flux, but also the robustness of deviations of the inversion-derived fluxes from the prior (and their significance given the uncertainties).**

  We have improved Figure 5 by adding prior estimations and prior errors bars for the different regions. It gives a direct indication about the deviations of our different inversions from the prior. Moreover, in a grid-point-scale variational system, like the system we used here, the computation of posterior uncertainties is highly time consuming. In the revised version, we report the posterior uncertainties provided by Cressot et al. (2014) for an inversion configuration, which is very close to ours (similar observation data sets, similar prior covariance matrix, similar optimization algorithm, LMDz model, etc.). In the updated text and according to reviewer's comment, 1/ we give the posterior error bars for BG-TD, EXT-TD and PR-LEI-TD inversions on the Figure 5, 2/ we propose a discussion on the significance of uncertainties due to parameterization errors given the posterior uncertainties in each region.

- **5. Further information is needed about the treatment of the initial concentration and the atmospheric oxidation in the inversion. Are they optimized? If not, could an inconsistency between the initial concentration field at the start of the short-window satellite inversion compared to the longer window surface inversion explain differences in the derived global total? How about the global sink?**
  In our system, we optimize the weekly grid-point emission flux of CH_4 together with the initial conditions of CH_4 mixing ratio (in the form of 2-D scaling factor on the CH_4 columns). The bias between measured and optimized surface concentrations is explained both by a modification of the initial conditions and by a modification of the methane emissions. Here, we found a bias of 40 ppb at the surface after the satellite inversions (see Figure 2). After analysing the scaling of the optimized initial condition, we found that around 15-20 ppb of the 40 ppb are explained by an increase of the initial condition, the rest being explained by an increase in the CH_4 emissions.

OH and $O(1D)$ fields are prescribed (optimized in the state vector but with a small error bar of 1%). These fields come from a full-chemistry simulation of LMDz-INCA (Szopa et al. (2013)) and after a global scaling by methyl-chloroform observations made before inversions. The different characteristics of the OH field (global mean concentration of 11.5 x 10^5 molec.cm^{-3} between surface and 100 hPa) are in the range of the current knowledge on the radical hydroxil (see the ACCMIP experiment ; Naik et al. (2013), i.e. between 7.4 and 13.3 molec.cm^{-3}). No inter-annual variability is applied to the OH field used here. We added more details on this in the updated text.

- **6. Even if the oxidant fields are the same, the lifetime may be different due to differences in transport.**
  We agree that the lifetime may be different due to differences in the modelling of atmospheric transport. We quantify it in Locatelli et al. (2015) (see their Figure 10). They found that differences in CH_4 lifetime simulated by the 3 versions of LMDz could reach 0.2 years. Indeed, they found a difference of 25 ppb in the state equilibrium of CH_4 mixing ratio simulated by LMDz-TD and LMDz-NP, after two 39-years simulations.
  We now give more information in the text about the treatment of the initial concentration and the atmospheric oxidation in the inversion.

- **7. From the results it is clear that some representations of transport are more realistic than others. It would be interesting to know of this translates into optimized models that are more or less realistic. The comparison with HALOE in figure 3 is clear, but it is unclear whether improved performance can also be demonstrated in the troposphere – which might relate more directly to the accuracy of the inversion-derived fluxes.**
  Locatelli et al. (2015) investigated the skills of the different versions of LMDz

in the troposphere and discussed the implications for inverse modelling. In particular, LMDz-NP improves the representation of PBL dynamics through the use of the thermal plume model. Large-scale atmospheric processes are better represented in versions using the deep convection scheme of Emanuel (1991). Then, we can expect more realistic inverted fluxes when using LMDz-NP and LMDz-AR4. We refer more clearly to this former paper in the revised version of the manuscript.

Specific comments

- **Page 11862, line 12: "Monteil et al, 2013"**
  Yes, we have included this reference.

- **Page 11864, line 5: Has the HALOE dataset been corrected for the CH_4 increase since those measurements were made?**
  No, the HALOE dataset has not been corrected for the CH_4 increase. Indeed, we are only interested here on the CH_4 gradient in the UTLS (Upper Troposphere/Lower Stratosphere) region, which is less sensitive to atmospheric increase than the absolute mean value.

- **Page 11864, last paragraph: I do not understand the second step of the inversion. In the second step the bias is quantified at each surface side, but how is that use in the second inversion step?**
  After a first inversion using GOSAT data, we compute the difference at each surface station between the CH_4 surface measurements and the simulated CH_4 mixing ratios based on the optimized flux coming from the first inversion. It

quantifies the consistency between surface and satellite inversions. In our case, we found a positive latitudinal bias (about +40 ppb) between simulated CH_4 mixing ratios sampled at the surface and CH_4 surface measurements. It means that surface and satellite data are inconsistent.
We consider that surface measurements are unbiased and we correct the satellite data accordingly using a latitudinal correction before performing a second inversion. In the paper, we suggest that most of this bias may come from the satellite data as LMDz-39 clearly improves the troposphere-stratosphere exchange compared to LMDz-19, which had only 19 vertical levels and only a 4 ppb bias with surface observations.

- **Page 11865 first paragraph: How is the global sink treated in the inversion?**
  We use a prescribed OH field coming from a full-chemistry LMDz-INCA simulation (Szopa et al. (2013)). We detailed it in the "general comments" section.

- **Page 11866, line 6: This could be, but it depends on where the surface measurements are made (it would not be the case e.g. for SPO).**
  Yes, we clarified this point in the text.

- **Page 11867, line 19: How do you define IH gradient here? Should not it rather be called "hemispheric difference"**
  Yes, it is the difference between CH_4 concentrations from the Northern and the Southern hemisphere. We change IH gradient into hemispheric difference in the text.

- **Page 11868, line 20: This conclusion is very sensitive to the relative weights of different measurement datasets in the inversion. If the weight of**

GOSAT is less than that of the surface network, that may also explain why the transport parameterizations have less impact on the fluxes.
We have based our inversion set-up on the work of Cressot et al. (2014), who have largely studied and optimized the error statistics of surface and satellite inversions. Consequently, we think that the GOSAT satellite data and the surface measurements are quite properly weighted in our system with respect to their own uncertainties.

- **Figure 3: Do the model contributions to the total column account for the averaging kernel of the satellite retrievals? This should be made clear.**
  We computed model contributions to the total column without accounting for the averaging kernel of the satellite retrievals. We specify it now in the legend of the Figure 3.

Technical corrections

- **Page 11856, line 1: "SCIAMACHY" i.o. "SCHIAMACHY"**
  Done.

- **Page 11857, line 4: "surface" i.o. "surrface"**
  Done.

- **Page 11859, line 28: "presented" i.o. "presenteed"**
  Done.

- **Page 11864, line 25: "methane flux" i.o. "methane fluxe"**
  Done.

- **Page 11868, line 20: "that" i.o. "than"**
  Done.

- **Page 11871, line 20: "span" i.o. "explore"**
  Done.

- **Table 2, caption: "shown" i.o. "showed"**
  Done.

- **Figure 4, caption: "institute" i.o. "institude"**
  Done.