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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. In general the discussion would benefit from a clearer analysis and separation of the two major sources of error which were identified in the introduction: observations vs. transport errors. Of course representation error kind of mixes up these two categories, but for the purposes of this study the two have been effectively separated. When I look at Figure 5 it seems that for these large regions it is often the case that the three different observing systems cause a spread as large or larger than what is seen for the same observing system with three different version of physical parameterizations (i.e. the difference between the three reds is as big or bigger as the difference between the red, blue and green for each region). The material is there to clearly describe and define this, but the discussion of this point is lacking. An improvement of this point would benefit the manuscript overall.**

  We agree with this remark of the reviewer. Accordingly, we added Figure 6, which compares the spread in inversions due to the choice of the measurement dataset (blue error bars) and the spread due to the choice of the version of the model for each region (green error bars). In red, we represent the spreads of the 9 inversions. These spreads are plotted as a minimum-maximum range.
  In few regions, the spread between inversions using different version of the model and inversions constrained by different datasets are similar (examples: South America temperate, Africa, Australia and Boreal Eurasia). However, we notice that the spread found in inversions using different datasets are much larger than the spread found in inversions based on different versions of LMDz for several regions, such as South America Tropical, Europe and China.
  Consequently, we discuss these different results in the paper in the Section 5.2.

- **2. What was missing in this study was a discussion of the sinks of methane. I read it quite carefully, and I'm not entirely sure if the OH sink was being optimized (let alone the soil sink, or if the Cl sink was even considered).**
  In this study, OH and $O(^1D)$ fields are prescribed (with a very small error bar of 1%). They are coming from a full-chemistry LMDz-INCA simulation of Szopa et al. (2013). The different characteristics of the OH field used here are in the range of the current knowledge on the hydroxyl radical exposed in Naik et al. (2013). Besides, reactions of CH_4 with chlorine are not considered in our system. We precise more clearly these points in the updated version of the text also acknowledging that the focus is more on methane emissions than on methane sinks.

- **3. (...) If it was being optimized, it would be interesting to see how the vertical mixing affected the magnitude and location of the tropospheric methane loss. If it is not being optimized, the differences in vertical mixing likely impact the lifetime simulated under each version of the model, and thus the global fluxes shown in Figure 4. In any case, it needs to be explicitly discussed.**
  As also stated in the answers to reviewer #1 comments, the different versions of LMDz derive different methane lifetime. We studied the impact of the vertical mixing on the methane lifetime in Locatelli et al. (2015). We found that the difference in methane lifetime due to changes in physical parameterizations could reach 0.2 years. It has been shown that the version of LMDz using the thermal plume model (LMDz-NP) simulated a methane lifetime 0.2 years higher that the LMDz-TD and LMDz-SP versions. You can have a look at the Figure 10 of Locatelli et al. (2015), which shows the different CH_4 mixing ratio equilibrium

  states reached after several years of simulation. These equilibrium states differ up to 25 ppb. Consequently, the different representation of the vertical mixing in LMDz modifies the methane lifetime in the different versions of LMDz and it directly impacts the estimation of methane emissions by inverse modelling. It is one contribution of transport errors leading to the uncertainties in inverse modelling.
  We specify now this point in the Section 2.2, referring more clearly to our previous paper on this important matter.

- **4. Although many numbers are used to describe the differences, the reader is left unsure about how significant an effect this is. Having the mean spread over several years for the surface-based inversions (in Table 2) is a start, but it doesn't show whether the patterns are consistent over these years, or whether the differences are more random in nature. Having only one year analyzed for GOSAT inversions exacerbates this. Although it might be significant extra work, considering the uncertainty on the posterior flux estimates would be an appropriate way to address this.**
  As mentioned by the reviewer (and also by reviewer #1), the computation of posterior uncertainties is very time consuming in such a large variational system. However, here, we can benefit from the study of Cressot et al. (2014), who have run Monte-Carlo simulations to compute posterior uncertainties for inversion configurations that are very close to ours (similar observation data sets, similar prior covariance matrix, similar optimization algorithm, etc.), and with the same transport model LMDz. In the revised version of the manuscript, we have applied the uncertainty reductions found by Cressot et al. (2014) to the results of our study. On Figure 5, we plot now the posterior error bars for BG-TD, EXT-TD and LEI-TD inversions, which correspond to the uncertainty reductions of Table 2 of Cressot et al. (2014).

We propose now in the text a discussion on the significance of uncertainties due to parameterization errors given the posterior uncertainties in each region (see Section 5.2).

Minor concerns

- **1. What is used for the driving meteorology? ERA-interim? I could not find this information easily in the paper. If ECMWF driving meteorology is used, did you consider using the convective mass fluxes that are stored? This is more consistent with the underlying transport of the model, which might solve some of the interhemispheric gradient problems associated with inconsistent schemes used to address sub-gridscale convection.**
  LMDz is a GCM and therefore it computes its own meteorology. In order to be more realistic, as classically done in many models, we nudge the LMDz horizontal components of the wind towards analysed winds from ERA-Interim. We then archive all the air mass fluxes and computes the inversion with an offline version of LMDz. Therefore the consistency is garanteed between meteorology and tracer transport.
  We specify more clearly these points now in the text (section 2.2).

- **2. The reference to GrooB and Russel in the text states that it's from 2014, but it's actually from 2005. But more importantly, details are missing with**

respect to how the comparison was carried out. Were the data corrected to account for trends in methane between 1991 (the beginning of the period used to compute the HALOE climatology) and 2010?
We have corrected the reference to GrooB and Russel.
The HALOE data have not been corrected for trends in methane between 1991 and 2010. Indeed, here we are mostly interested in the CH_4 gradient in the UTLS (Upper Troposphere/Lower Stratosphere) region. Therefore, we consider that correcting CH_4 concentrations is not important as we focus on the large differences in CH_4 mixing ratio in the UTLS between model versions, which are not very sensitive to the mean atmospheric value. For instance, difference between CH_4 mixing ratio simulated by LMDz-39 and LMDz-19 reach 500 ppb at 10 hPa! We clarified the text on this point.

- **3. Furthermore, is the model subsampled in a way consistent with the measurements (in terms of space and season)? HALOE didn't measure much at high latitudes ($\geq$ 50 degrees or so), where stratospheric methane is particularly variable. Was this taken into account? Why not use a more modern sensor such as MIPAS or ACE-FTS in addition (or instead) ?**
  Yes, the model has been sampled at the same location and time that the HALOE data. For the comparison, we only use data located between 60 $^\circ$ N and 60 $^\circ$ S for the whole year 2010. Moreover, we have not used others sensors because we consider that the results shown with the comparison using HALOE data are clear enough to support our point about the improvement from LMDz-19 to LMDz-39 regarding UTLS exchanges.

- **4. I am also slightly confused by what is shown in the "percentage" profiles in Figure 3. Is this the contribution of each of the GOSAT retrieval layers? And if so, for an average of all columns for 2010? Or something else? This**

needs to be better explained. Although chronologically in the manuscript it might be hard to work in, I was wondering what the different versions of the LMDz-39 looked like on this plot. Perhaps it would be instructive to include a similar comparison, perhaps for zonally-averaged columns in the tropics, NH extra-tropics, and SH extra-tropics. This might work well in a discussion of the photochemical sink, and how that effects the estimated lifetime across model versions (see comment above).

Yes, this is the contribution to each retrieval layer to the total column (in percentage), which is shown on the Figure 3. It has been plotted to show that stratospheric concentrations contribute more to the total column in LMDz-19 than in LMDz-39. On the contrary, tropospheric concentrations contribute more to the total column in the LMDz-39 version. This is directly related to the vertical profile shown on the right side of the Figure. We have clarified the text in the updated version.

The other versions of LMDz with 39 layers (LMDz-SP and LMDz-NP) have a very similar vertical profile to the one shown (LMDz-TD) on this plot. LMDz-TD, LMDz-SP and LMDz-NP may have large differences in the simulation of the vertical profiles at some specific location and time, but in the case of an annual global mean (as is shown on the Figure 3) the three versions of the model are very similar. It is confirmed by the Figure 2 where we show that the bias between surface measurements and surface optimized concentrations are very similar in the three 39-layer versions of LMDz. So, we have decided to show only one version of 39-layer LMDz here (the LMDz-TD version), which is consistent with the LMDz-19 version because they both use the same physical parameterizations. We precise this agreement for Figure 3 in the updated version of the text.

- **5. To be honest, I am surprised that the transport differences do not result in larger flux discrepancies in Figure 4. How do these differences**

compare to the posterior uncertainty? Is this something that your system can easily calculate? This question arises again when looking at Figure 5. How significant are the differences between the different implementations of transport? Do they result in posterior flux estimates that do not have overlapping uncertainties? The information to judge this is not provided. A 5% range due to transport differences is significant if the uncertainty is 1%, but not if it's 4%. Was the lifetime/OH sink fixed between simulations?

First of all, please remind that only transport differences due to the different physical parameterizations implemented are considered here. Differences derived in estimated methane emissions due to the modelling of atmospheric transport were much larger in Locatelli et al. (2013), where we use different models (different resolutions, different parameterizations, different analysed winds, etc.). Then, we were not surprised to find a smaller spread in this study than in Locatelli et al. (2013).

In order to quantify the spread in inversions due to differences in parameterizations relatively to posterior uncertainties, we use the uncertainties reductions found in Cressot et al. (2014) as explained before in this review. Thus, on the Figure 5, we give the posterior error bars for BG-TD, EXT-TD and LEI-TD inversions, which correspond to the Table 2 of Cressot et al. (2014).

We also propose a discussion (see the Section 5.2) on the significance of the impact of parameterizations errors on inversions relatively to the posterior uncertainties. As stated before (general comment #2), the OH field was prescribed based on a MCF calibrated field coming from the full chemistry model LMDz-INCA (Szopa et al. (2013)).

Typos/language issues

- **p11854, line 15: total-column what? total-column abundances, or total column methane mixing ratios, etc., something is missing there.**
  Ok, we clarified it in "total-column mixing ratios".

- **p11854, line 18: gradient → gradients**
  Ok.

- **p11855, line 1: relatively → relative**
  Ok.

- **p11855, line 12: supplement the issue? Or rather ameliorates the problem? Or they supplement the existing measurement network...**
  We decided to use "solve the issue".

- **p11855, line 14: become → becomes. Also, it was already a major issue, perhaps now it becomes the leading issue?**
  Ok.

- **p11855, line 19: satisfactory → satisfactorily**
  Ok.

- **p11856, line 1: SCHIAMACHY → SCIAMACHY**
  Ok.

- **p11856, line 5: carry on → carry out**
  Ok.

- **p11856, line 5-6: have also → also have**
  Ok.

- **p11859, line 19-20: "by Tiedke (1989) scheme" → "by the scheme from Tiedke (1989)" or "by the Tiedke (1989) scheme", similar with Yamada**
  Ok

- **p11859, line 24: "by Emanuel" → "according to Emanuel" or similar**
  Ok.

- **p11859, line 27: an → a**
  Ok.

- **p11860, line 5: "On the opposite" → "On the other hand"**
  Ok.

- **p11860, line 6: "has been also" → "has also been"**
  Ok.

- **p11860, line 7-10: Rework the sentence a bit. Perhaps: "The interhemispheric (IH) exchange, which is known to be too fast in LMDz-TD, agrees better with the indirectly measured IH exchange when using the Emanuel (1991) scheme, as is done in LMDz-SP and LMDz-NP.**
  Ok, done.

- **p11860, line 11: "which justify to test it as well" → "which justifies its inclusion"**
  Ok.

- **p11861, line 19: "that CO2" → "that the CO2"**
  Ok.

- **p11863, line 22: "that CH4" → "that the CH4"**
  Ok.

- **p11864, line 11: "Consequently, the inverse system derives lower methane fluxes with LMDz-19 to simulate lower tropospheric methane mixing ratio compensating the over-contribution of stratospheric methane mixing ratio to the total-column." → "Consequently, the inverse system derives lower methane fluxes with LMDz-19 to simulate a lower tropospheric methane mixing ratio, compensating the over-contribution of the stratospheric methane mixing ratio to the total-column."**
  Ok.
- **p11864, line 18: "modelling of" → "modelling of the"**
  Ok.
- **p11864, line 19: "reasons of" → "to determine the reason for", "need" -> "needs"**
  Ok.
- **p11864, line 25: fluxe → fluxes**
  Ok.
- **p11864, line 28: "we only focus and present results associated to " → "we focus on and present only results associated with "**
  Ok.
- **p11865, line 10: "which was estimated as a “total” transport model errrors" → "which was an estimate for "total" transport model errors"**
  Ok.
- **p11865, line 13: "although smaller than" → "although a smaller impact than"**
  Ok.

- **p11865, line 23: "on China methane flux estimates" → "on the methane flux estimates for China"**
  Ok.
- **p11866, lines 1 and 4: "simulated total-column" → "the simulated total column"**
  Ok.
- **p11866, line 8: "total-column" → "the total column"**
  Ok.
- **p11866, line 17: "have been" → "has been"**
  Ok.
- **p11867, line 23: "wrong repartition between Northern and Southern Hemisphere of emissions" → "incorrect repartitioning of emissions between the Northern and Southern Hemispheres"**
  Ok.
- **p11868, line 1: southern → Southern**
  Ok.
- **p11868, line 5: extra-tropics → the extra-tropics**
  Ok.

- **p11868, lines 10-11: reach 7.5 unitTg CH4 year**
  Ok.

- **p11868, line 14: impact strongly → strongly impacts**
  Ok.

- **p11868, line 20: than → that**
  Ok.

- **p11868, line 21: impacts → impact**
  Ok.

- **p11869, line 19: LMDz-SP and LMDz-SP → I guess this should be LMDz-SP and LMDz-NP, right? and also "the Emanuel"**
  Ok.

- **p11869, line 21: dependent ON**
  Ok.

- **p11869, line 23: "Then, LMDz-SP and LMDz-NP derives also" → "Thus LMDz-SP and LMDz-NP also derive"**
  Ok.

- **p11871, lines 6-7: "where modelling of boundary layer mixing impact much atmo- spheric methane levels" <- I'm not entirely sure what is meant here,**

please reword it. Does boundary layer mixing have a large impact on the concentration of atmospheric methane? Or does boundary layer mixing impact the atmospheric methane concentration across several model levels?
Here, I explain that stations added in the EXT configuration are located closer to methane sources. Ok.

- **p11871, line 16: are ranged from → range from**
  Ok.

- **p11871, line 27: deriving → derive**
  Ok.

- **p11873, lines 4-7: "Indeed, inversions using Emanuel (1991) scheme (based on LMDz-SP or LMDz-NP model) have smaller interhemispheric methane emission gradients than inversions using Tiedtke, 1989, scheme (based on LMDz-TD model), which are known to simulate too fast inter-hemispheric exchange (Patra et al., 2011)." → "Indeed, inversions using the Emanuel (1991) scheme (LMDz-SP or LMDz-NP) have smaller interhemispheric methane emission gradients than inversions using Tiedtke (1989) (LMDz-TD), which are known to overestimate interhemispheric exchange (Patra et al., 2011)."**
  Ok.

- **Figure 3, caption: profils → profiles**
  Ok.

- **Figure 4, plot: Physic → Physics; subscript of 4 in CH4**
  We redo the Figure 4 taking into account your comment.

- **Figure 4, caption: change "Leicester institute", remove comma after "and" (or move it before)**
  Ok.