

Response to referee #1

*Major comments:*

1. *The paper is focused on model evaluations. But no necessary detail on these models such as the surface CH<sub>4</sub> fluxes, meteorological fields, model resolution, and chemistry scheme are presented (although references are provided), or are used to explain their different performances (for example Figure 3).*

All the three models optimize CH<sub>4</sub> field against in situ measurements at the surface through inversions of the CH<sub>4</sub> emissions. The chemical reactions considered in the models are the oxidation by OH in the troposphere, and by Cl, OH and O(<sup>1</sup>D) in the stratosphere. The fields of the radicals are prescribed monthly with no interannual changes. A description added in the first paragraph in page 5 to illustrate those. The meteorological fields, model resolution are included in Table 2.

2. *Biases in GOSAT retrievals, and their implications on the model evaluations have not been discussed by the authors. GOSAT XCH<sub>4</sub> has not been fully validated (particularly) over tropical regions, and could itself have latitude-dependent bias as well. I suggest the authors use more recent version of GOSAT XCH<sub>4</sub> retrievals (such as OCPR v7) as well.*

We follow the advise of the referee, and the version OCPR v7 of GOSAT data is used in the Figure 2.

*Minor comments:*

1. *Line 37, Page 1: ‘... 6.2 +/- 11.2’ ppb in the stratosphere ... ’ The notation of +/- 11.2 ppb may be mis-leading, as in this case the ‘amplitude’ as defined by the authors could not be negative.*

The symbol '+/-' has been changed to '±' in the text.

2. *Figure 1: Caption and main text does not provide necessary information, for example, the information about IMECC, and aircore data etc.*

The FTS data are averaged for the in situ measurement periods. The IMECC is an aircraft campaign over Europe (Geibel et al., 2012). The Lamont-AirCore measurements are from Greenhouse Gas Group Aircraft Program (<http://www.esrl.noaa.gov/gmd/ccgg/aircraft/>). The AirCore data at Sodankylä is from the FTS group there. Following this advise, the information has been added to the caption of Fig. 1.

3. *Line 6, Page 4: ‘... infers dry air columns from the CO<sub>2</sub> columns retrieved from the same spectra as used in the CH<sub>4</sub> retrieval’ The sentence is not clear, and no mention of model CO<sub>2</sub> concentrations, which is one of the possible sources for biases in GOSAT proxy XCH<sub>4</sub> data.*

The sentence has been changed to ' .... spectra as used in the CH<sub>4</sub> retrieval. This method assumes the CO<sub>2</sub> concentrations are known and provided by model simulations. ' at line 16 in page 4.

4. *Line 14, page 4: ‘... F07\_10 data are applied and measurements with less than 1.4 DOFS are filtered out...’, More detailed information such as the observation coverage and errors will be helpful.*

Validation of F07\_10 data against to HIPPO measurements shows a bias of -8 ~ 5 ppb with standard

deviations of 25 ~ 50 ppb below 100 hPa (Herman and Osterman, 2014). According to the advise of the referee the information has been added to the line 24~25 in page 4.

5 5. Line 30, Page 5: '*... Figure 3 shows yearly and seasonal median model biases scaled by the fraction of the air column in the troposphere and stratosphere ...*'. I suggest adding the number of the TCCON observations at different months to the plot. Also it is interesting to know whether TCCON retrievals have biases depending on the solar zenith angles.

10 There are 10 sites used in that plot, for all seasons, except for measurements at ZEP is absent during the season DJF and SON. This is clearly seen in Fig. 3. The TCCON products has been corrected for solar zenith angle dependence. So this bias should be minor.

15 6. Line 5, Page 6: '*... one can see that the latitudinal pattern of model biases in total column-averaged CH<sub>4</sub> results from both the stratosphere and troposphere for ...*' Some explanation of different performances of the three models shown in Figure 3 in terms of surface fluxes, transport or chemistry scheme will be helpful.

20 All the models are optimized with respect to surface measurements already. So the surface emission might not been the main reason to the different performances. The tropospheric oxidation also directly influence the surface CH<sub>4</sub> concentrations, and the optimization process should give emissions consistent with the prescribed OH field in each model. The only significant difference among the models could come from convection, the North-South transport, and the transport from the troposphere to the stratosphere. However, it is difficult to give some useful discussion since only  
25 column measurements are used to evaluate the models.

30 7. Line 20, Page 6: *TCCON and in situ sites are selected to be located close to one another so that both instruments measure similar airmasses ::: The TCCON and in-situ measurements have different measurement frequencies. For example, availability of TCCON data usually has strong seasonal variations. How will these differences affect the results presented in Figure 4?*

35 Yes, the TCCON measurement has a different sampling frequencies compared to the in situ measurement. In our analysis the measurement series of TCCON and in situ has been filtered to extract variations with temporal scale longer than 1.4 years and only multi-year (longer than 3 years) averaged results are used. Besides, the models are matched to TCCON and in situ measurements in time, respectively, and undergo the same analysis with the measurements. So the model performance against to the measurements should not be affected.

40 8. Table 3: typo: *The latitude of the Lauder TCCON site should be -45.038.*

We follow the advise, and this error has been corrected.

45

Response to referee #2

The comments by referee #2 mainly include two parts: 1) The tropopause applied to integrate the model  $CH_4$  to obtain tropospheric and stratospheric column-averaged  $CH_4$  should be checked and the results could depend on the definition of the tropopause. 2) The comparison of the model with TES measurements does not support our conclusions.

1) For the first comment some sensitive tests have been conducted, some different definitions for the tropopause are applied to integrate the model outputs. That include, thermal tropopause according to WMO definition, the dynamical tropopause defined as 1.5, 2.5, 3.5 and 4.0 PVU surface in the extratropics and 380 K potential temperature surface in the tropics. The ECMWF-interim reanalysis data is used to calculate the tropopause.

The sensitive test is applied to TM3 and LMDz-PYVAR (TM5 has a similar configuration with TM3 and then not been tested). These sensitive tests show (see following plots) that, the tropospheric model bias almost is not affected by the selection of the tropopause, even for the unrealistically low tropopause of 1.5 PVU. The amplitude of the stratospheric model bias changes between the thermal tropopause and the dynamical tropopause of 2.5~4.0 PVU. However, there is still not a consistent latitudinal gradient existing during whole year for the stratospheric model biases. So the conclusion that the latitudinal gradient in the model bias of total column-averaged  $CH_4$  come from the troposphere is valid. The dynamical tropopause of 1.5 PVU give some latitudinal pattern in the stratospheric model biases, but, that tropopause is unrealistically low and frequently reach 170 hPa (below 380 K potential temperature surface) in the tropics.

2) For the second comment, the results from TES actually support the conclusion that the inconsistency between the HIPPO and TCCON comparisons with the models come from the longitudinal dependence of latitudinal gradient in the tropospheric model bias. But there are writing errors in the figure caption of the Figure 6 in the manuscript. In the third panel of Figure 6, the black points correspond to HIPPO sampling area (110°W~150°E) and the red points to the region beyond it.

Additional comments:

1. As explained on page 6, biases are assessed by taking the absolute difference between model and FTS. The motivation is that biases may change sign seasonally, and therefore may not show up in annual averages when positive and negative contributions cancel out. However, whether this is a good choice or not depends on the kind of bias that is investigated. Here the focus is largely on a latitudinal bias. Suppose that there is no latitudinal bias in the annual mean, but only a latitudinally varying bias in the seasonal amplitude. By taking absolute model to FTS differences across the year you would end up with a latitudinally varying bias. In this case the choice of absolute differences was clearly not appropriate. There may not be a single solution to this problem for the biases that are investigated here, but the meaning of the numbers that are summarized in the abstract and the conclusions for stratospheric and tropospheric contribution to the bias is not clear to me. A relation with a latitudinally varying bias is suggested, but do these numbers really reflect stratospheric and tropospheric contributions to that bias. This requires more attention, including information on how the absolute differences are calculated (on every data point like an RMS, or on

monthly averages, or?).

5 The absolute difference between the models and FTS is only used to calculate the averaged bias over all years and all sites. The results of the absolute bias are only the numbers appearing in Page 6, line 29~30 and in the abstract. The true bias (model-measurements) is used for all other parts of the paper, including all the plots.

10 2. Looking at Figure 5, the most significant differences between the models and HIPPO seem really at the highest measured altitudes. You might debate whether they are in the troposphere or the stratosphere. I wonder how important this really is. Wouldn't it be better to conclude that the problems show up most strongly at tropopause altitudes. In that case the method of separating the troposphere from the stratosphere may actually not be so appropriate. A plausible cause could be strat-trop exchange. I don't see how the results that are presented here exclude this possibility. Yet, it is not considered as an option.

15 Yes, the model bias indeed increase abruptly above the tropopause. However, the approach separating the troposphere from the stratosphere does not influence the latitudinal gradient in the model biases of tropospheric CH<sub>4</sub> as show in the Fig. 1-5. Only the stratospheric model biases are sensitive to the separation method and appear large when the tropopause is defined as low as 2.5 PUV. But the stratospheric model biases did not present a consistent latitudinal gradient with the model biases in the total columns of CH<sub>4</sub>.

20

3. page 4, line 8: Where does the tropopause pressure come from?

25 In deriving tropospheric CH<sub>4</sub> from FTS measured total columns of CH<sub>4</sub> and N<sub>2</sub>O, the linear correlation existing between N<sub>2</sub>O and CH<sub>4</sub> in the stratosphere is applied. In the troposphere the N<sub>2</sub>O concentration is well known and then stratospheric N<sub>2</sub>O column is obtained through subtracting its tropospheric contributions from the total columns. Because of the correlation stratospheric CH<sub>4</sub> column is estimated from N<sub>2</sub>O columns and finally the tropospheric CH<sub>4</sub> column is known. In the process the tropopause pressure is not needed.

4. page 4, line 13: What model CO<sub>2</sub> fields are used to translate the retrieved ratios into XCH<sub>4</sub>?

30 The CO<sub>2</sub> field is from the CarbonTracker model.

5. page 5, line 13: 'The NCEP tropopause ...'. It is less accurate for TM5 also, which doesn't use NCEP either (in TM3 it depends on the meteo that was used). Please reformulate to make this sentence more accurate.

35 We redo the analysis with the thermal tropopause derived from ERA-Interim datasets. Now, the sentence has been changed to 'The thermal tropopause calculated using the reanalysis data ERA-Interim is used in all calculations, which could not be so accurate for the TM5 and LMDz models, especially for LMDz that predicts its own meteorology fields through nudging to reanalysis data.'

6. Page 7, line 18: 'underestimations dominate'. There are lower values elsewhere, so it is not clear that they 'dominate' in the SH.

40 The sentence has been changed to 'Underestimations dominate in the upper southern troposphere, consistent with the results in Fig. 4 that modeled gradients of tropospheric CH<sub>4</sub> are biased negative as revealed by FTS and surface measurements.'

7. *Figure 3: Please add vertical lines between the columns (i.e. models). At the boundary between the models it is not so clear which bar belongs to which model.*

We follow the advise of the referee, necessary modifications have been applied to the Fig. 5.

5 8. *Page 6, line 1: It would be fair to add Monteil et al, JGR, 2013 here, since they were among the first to report a latitudinal bias.*

The reference is added to the text on page 6, line2.

10 *Technical corrections:*

1. *page 2, line 4: 'transport' i.o. 'transports'*

2. *page 2, line 19: 'increase' i.o. 'incrase'*

3. *page 4, line 11: 'CH4' i.o. 'CO2'*

4. *page 4, line 11: 'applied to' i.o. 'applied from'*

15 5. *Page 7, line 2: 'except over' i.o. 'except for over'*

6. *Figure 4: the dashed zero line is missing in the upper panel*

7. *Page 7, line 23: 'show' i.o. 'gives'*

All the corrections has been incorporated into the manuscript except for the 6th. In upper panel of Figure 4 most of the values are smaller than zeros, so it is not necessary to draw a zero line there.

20

25

30

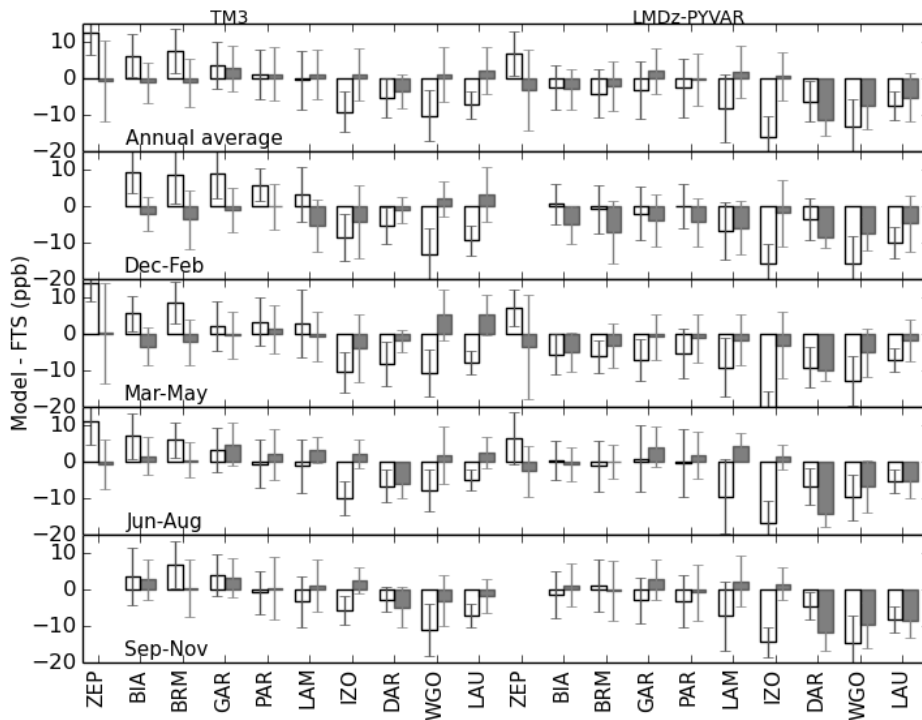


Figure 1. Yearly and seasonal medians of the scaled stratospheric and tropospheric contributions in modeled total column biases at TCCON sites. The sites from left to right is North to South. The white bar denotes the tropospheric bias, the grey bar for the stratospheric bias. The scale factor for the model bias are the air column fractions  $P_i/1000$  (stratosphere) and  $(1-P_i/1000)$  (troposphere), where  $P_i$  is the thermal tropopause pressure. The error bar are the standard deviations of the model biases. The results are averaged for 2007-2011 when FTS measurements are available.

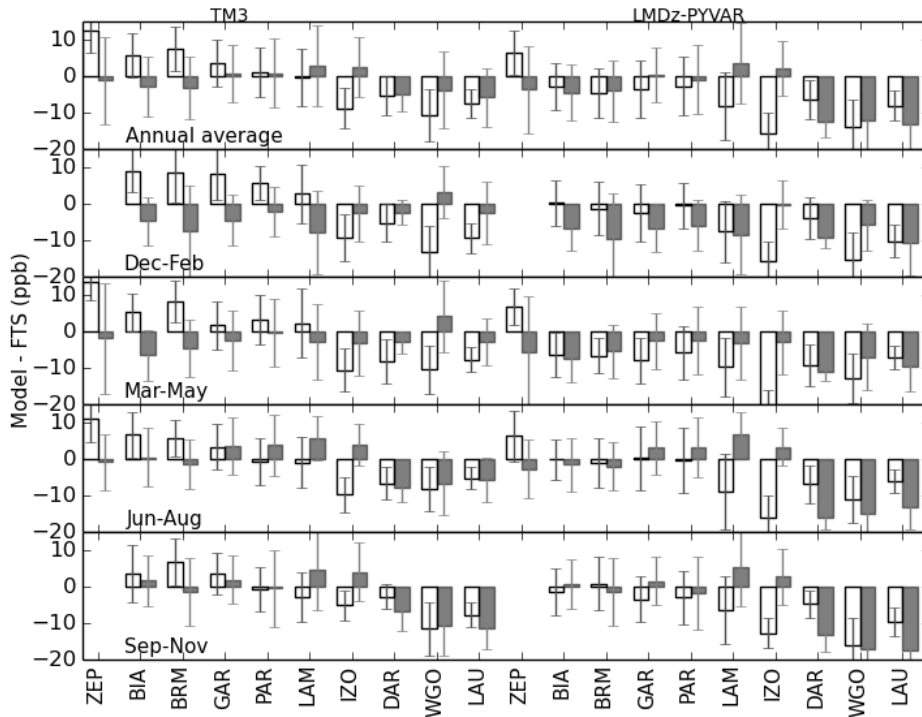
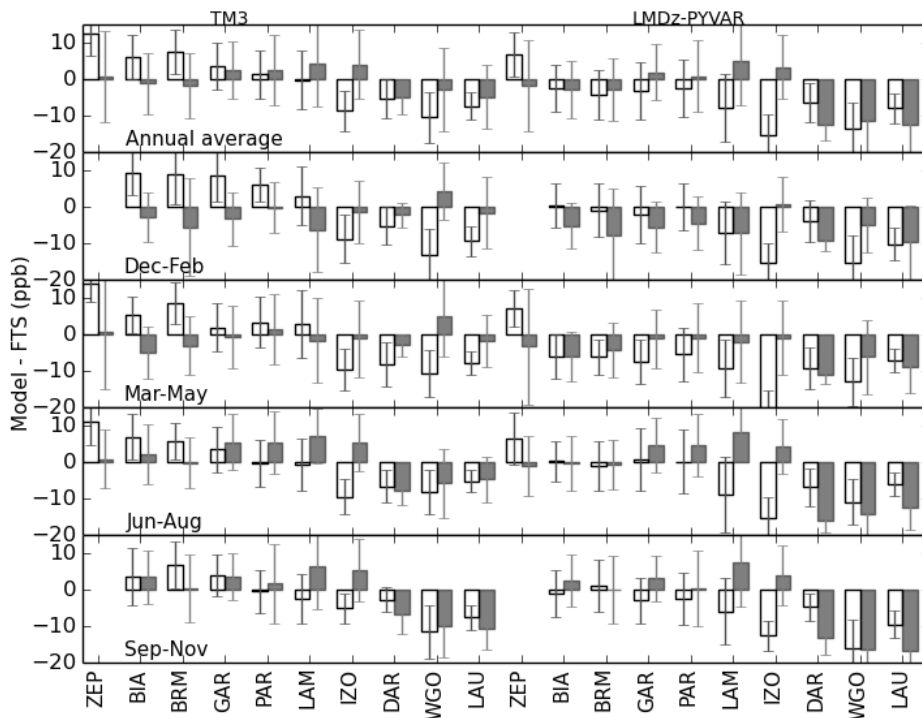


Figure 2. Same as Fig. 1 except for 4.0 PUV dynamical tropopause is applied.



5 Figure 3. Same as Fig. 1 except for 3.5 PUV dynamical tropopause is applied.

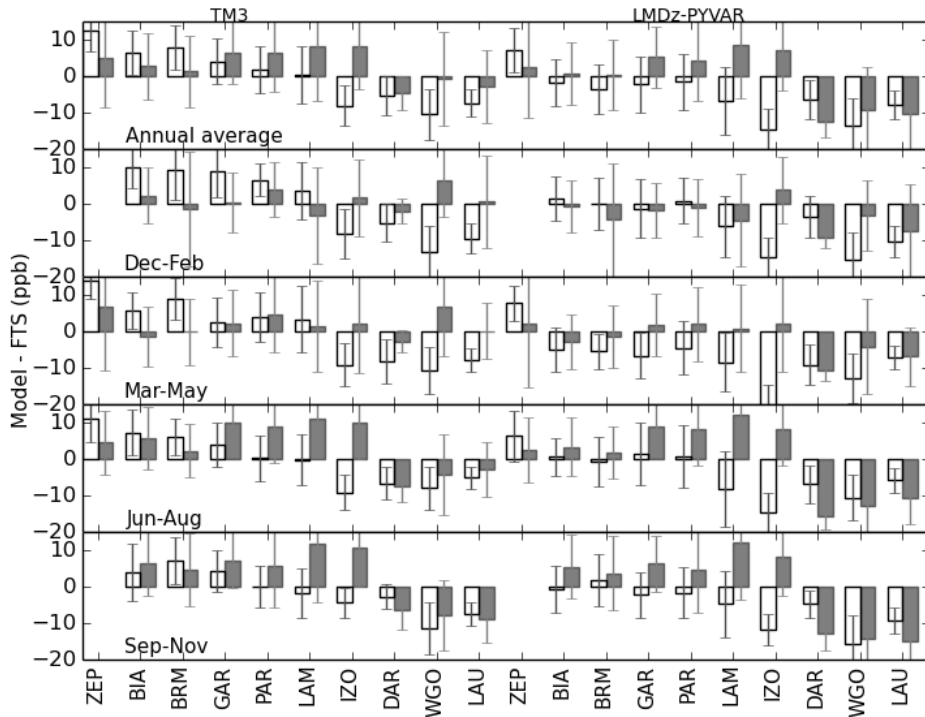
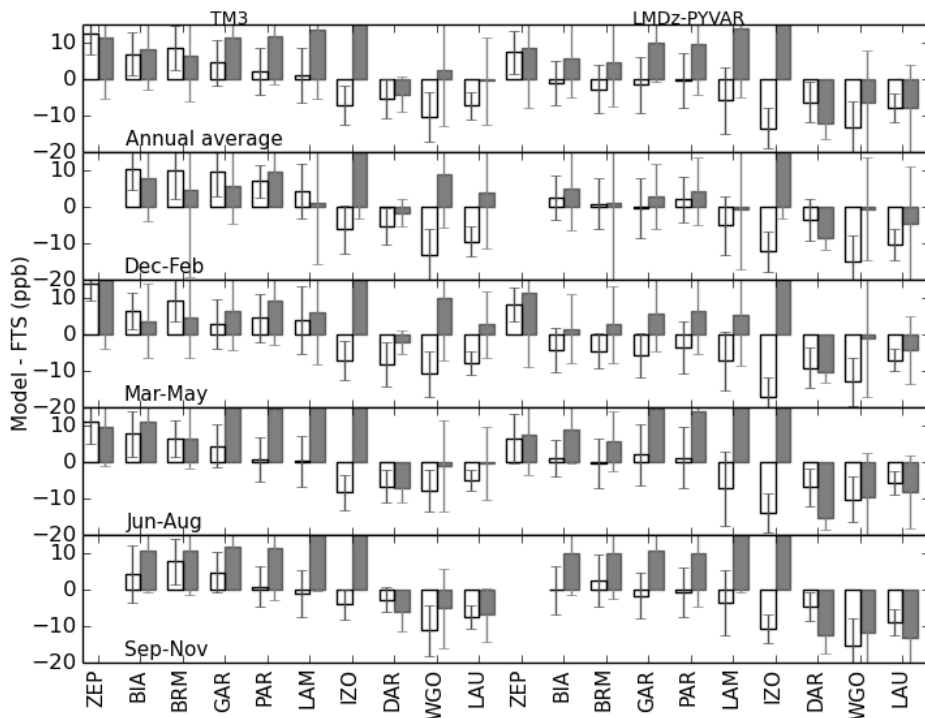


Figure 4. Same as Fig. 1 except for 2.5 PUV dynamical tropopause is applied.



5 Figure 5. Same as Fig. 1 except for 1.5 PUV dynamical tropopause is applied.