Response to Reviewer # 2

Anonymous Referee #2

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1 General Remarks

The paper compares columns of long lived trace gases simulated by a CTM driven by reanalysis data with observations. A new coordinate system in the model is introduced. Unfortunately the text is often not unique because of sloppy expressions or missing definitions. Uncertainties due to the use of climatologies for chemical sinks and precalculated heating rates are not enough addressed. Several Figures and Tables cannot be understood from the captions, but only after carefully reading the text (even that is not sufficient for some). The paper needs a lot of clarifications to be acceptable.

We thank the reviewer for their constructive and helpful suggestions. We have provided our responses to the reviewers' comments and believe our manuscript is much improved as a result. The reviewer's specific comments (shown in **bold**) are addressed below.

2 Specific Comments

Abstract: Give full expression for XCH4 and XCO2. Is this surface mixing ratio (like in other papers) or the quantity in the title? Please give locations and some reasons for discrepancies already there instead of words like 'bias' or 'contrasting performance'.

Here XCO_2 and XCH_4 are column-averaged dry air mole fractions of atmospheric carbon dioxide and methane, as in other papers.

"The simulated XCO_2 and XCH_4" is replaced with "The simulated column-averaged dry air mole fractions of atmospheric carbon dioxide (XCO_2) and methane (XCH_4)..."

Locations and reasons of discrepancies between simulated results and observations are described in detail in the text, e.g. section 3.2-3.4. In the abstract, we have given only more common results to outline a content of the paper.

Introduction:

Paragraph 3: I suppose 'column averaged' implies a pressure weighting and the quantity is related to the total column as seen for example by nadir viewing satellites. Please give a more detailed definition.

In atmospheric physics, total column density or total column is the mass of substance per unit area integrated along a path, typically a line of sight or from the bottom of the atmosphere to the top. Furthermore, if the total mass of dry air optically traversed is established, one may recover column-averaged concentrations (Dufour et al., 2004).

At present moment "column averaged" and "total column" are well-established definitions (e.g. O'Brien and Rayner, 2002; Rodgers and Connor, 2003), so more detailed explanations are not required in the paper.

Paragraph 4: CH4 is oxidized by OH and Cl mostly in the troposphere. The stratosphere has only a minor effect on surface mixing ratios. The total column as referred to in the reference is indeed dependent on tropopause height because of the decrease in mixing ratios above the tropopause. However, it is also dependent on the surface elevation which is not mentioned. Please reword paragraph to avoid misunderstandings.

We agree with the reviewer that CH_4 is oxidized by OH and Cl mostly in the troposphere. However in this Paragraph we emphasized that the CH_4 sink in the lower stratosphere is enhanced in comparison with the upper troposphere due to oxidation by O(1D). As result, the effects of variations in tropopause height are more pronounced for CH_4 than for CO_2 due to the contrast in CH_4 concentration above and below tropopause.

Dependence on the surface elevation can be considered easily, but more important is the variability over time, referred to in this section. To avoid misunderstandings we reworded:

"The variabilities (synoptic, seasonal, and latitudinal) in XCO_2 and XCH_4 are driven mainly by changes in …" with "The synoptic and seasonal variabilities in XCO_2 and XCH_4 are driven mainly by changes in surface pressure, …"

Paragraph 7: A lot of the problems discussed here arise from artifacts in the ERA 40 reanalysis.

The discussed problems arise not only for ERA 40 reanalysis, but they are known for other reanalysis (Hall et al., 1999). Key weaknesses of reanalysis are:

- The changing mix of observations, and biases in observations and models, can introduce spurious variability and trends into reanalysis output.
- Observational constraints, and therefore reanalysis reliability, can considerably vary depending on the location, time period, and variable considered. Changes in the observing system can also cause changes in mean errors.
- Mixing observations with models tends to violate conservation properties.

Effects of these issues are especially noticeable in the stratosphere, because the errors are comparable with the intensity of the stratospheric circulation.

Section 2.1: Please define all variables used in equation 1 and give the region where the transition is applied. Refer to Table 1.

Definitions of all variables used in equation 1 are added as follows: ζ denotes the level of the sigma–isentropic grid as described in Table 1, P and P_s are atmospheric pressure and surface atmospheric pressure respectively, $\theta = T\left(P_S/P\right)^{\left(R/c_p\right)}$ is potential temperature, T depicts temperature, R is the molar gas constant, c_p is the specific heat for a constant pressure, σ_θ and P_θ are "sigma" and pressure at the level θ_T, respectively. Transition region is the level $\theta_T = 350K$. Above θ_T grid is fully isentropic, below this level grid is terrain-following.

Section 2.2:

Paragraph 1: The use of precalculated heating rates based on climatologies of radiatively active gases (CO2, ozone, CH4) and the meteorological reanalysis introduces errors in the vertical transport due to inconsistencies. Especially ozone and its radiative interaction with clouds can be critical in the tropical lower stratosphere, but also in high latitude spring (ozone hole!). Some error analysis should be given.

Heating rates at the top of atmosphere are calculated by General circulation models (GCMs) using radiative transfer models and are partially constrained by satellites observations. The current GCM radiation schemes could have inaccuracies of around 10-20% in their total radiative forcing of the long-lived greenhouse gases (Collins et al., 2006; Forster and Taylor, 2006). As long as radiative heating rate is not an observable parameter one can only try to use the tracer transport and tracer correlations for evaluation of the heating rate data. It is also difficult to separate the errors in heating rate from the errors in horizontal and vertical mixing

of tracers. We use a vertical profile comparison for validation. The rationale for using climatological heating rate instead of instantaneous (like in TOMCAT model by Chipperfield et al) simulated values comes from our targeted application to very long lived tracers like CH_4 and CO_2 with a lifetime in stratosphere of the order of a year or more.

The JRA-25 data used to drive the NIES TM has a systematic large negative temperature bias up to 2 degrees in the lower and middle stratosphere and a large positive temperature bias up to 5 degrees in the upper stratosphere. These biases are significantly reduced in a new radiation scheme implemented in JCDAS (Onogi et al., 2007), but the JCDAS period was not included in climatology base data.

Paragragh 5: 300K is in most cases in the troposphere, except near the poles where normally no ascent occurs (last paragraph). This 'fudge factor' is confusing. There is also a contradiction to Eq. 1.

There is a mistype here. "300 K" should be replaced with "350 K".

Section 2.3: Please mention how the reanalysis data are converted to the model grid. Refer to Table 1 earlier. How often are the heating rates calculated? Every timestep and at every grid point or just climatological as said in section 2.2?

Heating rates are interpolated at every time step at the every model cell of sigma–isentropic grid using original JRA-25/JCDAS data available with three hourly time step.

Page 8061, line5-6: "… the NIES model derives the climatological heating rate from long-term global atmospheric reanalysis (see Section 2.3), which is provided as the sum of short- and long-wave components." replaced with "… the NIES model interpolates the climatological heating rate at every meteorology data update step (3h) at every model cell of sigma–isentropic grid using 2D monthly distribution of atmospheric reanalysis heating rate (see Section 2.3)."

In Section 2.3 we added:

"The 2D monthly distribution of the climatological heating rate used to calculate vertical transport in the θ-coordinate domain of the hybrid sigma–isentropic coordinate is prepared from JRA-25 reanalysis data, which are provided as the sum of short- and long-wave components on pressure levels."

Section 2.5: The use of the old OH climatology by Spivakovsky et al., (2000) in the troposphere and 2D-models in the stratosphere without any interannual variation can cause deviations for CH4 at different sites. Cl from seasalt is not mentioned. The sinks should be implemented in a better way. There is now also EDGAR 4.2 available. The use of constant emissions after 2007 causes larger deviations from observations. It might be better to show results only to the end of 2007.

For the CH_4 simulation, an inverse model-adjusted flux was used, obtained by optimising the surface fluxes of CH_4 using the LMDZ model for the period 1988–2005 (Bousquet et al., 2006). The methane concentration calculated using slightly outdated fluxes was manually adjusted, after simulation, by the annual mean concentration at the South Pole. Thus, we corrected misfit due to interannual variation. Given the high uncertainty in global CH_4 emissions (range between 500 to 600 Tg yr−1 from inverse estimates and even higher from bottom-up methods) and the small number of alternative OH distributions in global transport models (Patra et al, 2011) we consider combination of LMDZ inverse model-adjusted flux + OH climatology by Spivakovsky et al., (2000) as well balanced, which was successfully applied for global methane simulations (Patra et al, 2011).

At present, about 80% of CH_4 is removed by the hydroxyl radical (OH) in the troposphere alone (Fung et al., 1991; Lelieveld et al., 1998). It is estimated that Cl removes just 3–4% of CH_4 (Platt et al., 2004; Allan et al., 2010). Thus, methane sink due to reaction with chlorine from sea salt is not mentioned, as its effect is minor in comparison with OH uncertainty.

To assess the model results we selected the period January 2009 to January 2011, as we have intended to use the modeled XCO_2 and XCH_4 for estimation of GOSAT retrieval performance over the areas not covered TCCON sites. It was successfully implemented by Oshchepkov et al. (2012).

Section 3.3.1, paragraph 1: The bias should be due to wrong emissions and/or sinks. Fudging is no solution.

We are afraid there is misunderstanding in your comment regarding bias correction. To avoid misunderstandings, we decided to add the following sentences after the fifth Paragraph in section 3.4:

"Matching the model's mean CH_4 with the observations is achieved by adjusting either global total emissions or sinks, which both have large uncertainties (10-20%, Patra et al, 2011). Small residual offsets can be adjusted by tuning global emissions, but long-term simulations are required to reach and equilibration between sources and sinks. Adding a small 30 ppb offset to simulated results is nearly equivalent to the corresponding proportional change in the emissions fields on the order of 2%. For CO_2, the corresponding bias correction is about 0.5%."

Section 3.4:

Paragraph 3: Are the site specific profiles not seasonal? To include seasonal effects is very important at these sites. Is there a problem with the polar vortex at Sodankyla? This can easily explain biases by 5 to 10% as seen in a CCM which shows a clear anti-correlation between stratospheric CH4 column and potential vorticity at about 70hPa.

We believe it is very important to take into account seasonal effects at these sites. However, site specific a priori profiles are not available for this (current) TCCON release.

The reason for the large bias in February-April 2010 at Sodankyla site may be not only to the uncertainties in the model simulation, but also due to errors in FTS observation, which can be caused due to high zenith angle (a viewing geometry through high airmass). Sodankyla data are available for one year only, so it is very difficult to conclude there is a problem with the polar vortex. Moreover, Saito at al., (2012) also shown large bias between observed and modeled XCH$_4$.

Paragraph 5: Again troposphere and stratosphere are messed up. One reason for offsets might be indeed the not specific (or outdated) sinks in the troposphere and stratosphere.

Page 8073, line 21: "... the stratosphere." replaced with "... the atmosphere."

The main reasons that influenced on the simulation results are already addressed above.

Section 3.4.1, last paragraph: For most sites the variability of the model results is much less than for the observations. Especially at Sodankyla this appears to be due to the use of climatologies for calculation of vertical motions.

We disagree with the reviewer that it is possible to make such conclusion from comparison with TCCON observations. Moreover, the use of climatologies for calculation of vertical motions cannot affect the seasonal cycle of the model results, because a timescale of the stratospheric circulation variability is longer than the near surface seasonal cycle.

The size of the model grid cell is 2.5°×2.5° degree in horizontal resolution and ≥250 m in vertical direction. The model time step is about 15 min. In our simulation we use fluxes which also have quite coarse resolution (1.0°×1.0° degree). While every ground-based FTS TCCON

observation is measured in one stationary point in relatively short period. During measurements, many factors (local meteorology, local sources, clouds and others) may affect on the results. Therefore, XCO_2 and XCH_4 observed by ground-based FTS TCCON have higher variability than modeled ones even within one day.

3 Technical Corrections

Page 8058, line 29: height-coordinates!

We agree. "*z*-coordinates" was replaced with "height coordinates". But "*z*-coordinates" is also can be used (Kalnay, 2002).

Page 8070, line 17: Typo.

"Fig. 7" was replaced with "Fig. 6"

Page 8071, line 24: bad wording.

"subsides" replaced with "decreases"

Page 8072, line 21: Separate sentences.

A sentence

"To compare the modelled total column with measurements directly, it is necessary to consider the measurement averaging kernels those describe the sensitivity of the retrieved total column to a perturbation in absorber abundance in a given layer of the vertical profile (Rodgers and Connor, 2003; Wunch et al., 2011)."

is divided in two

"To compare the modelled total column with measurements directly, it is necessary to consider the measurement averaging kernels. Such averaging kernels describe the sensitivity of the retrieved total column to a perturbation in absorber abundance in a given layer of the vertical profile (Rodgers and Connor, 2003; Wunch et al., 2011)."

Page 8088, Table 1: remove horizontal lines in part on upper troposphere and stratosphere. The numbers in the table are only valid for an ocean surface. Say something on mountains in the caption or give p/ps =σ instead of p.

Horizontal lines in part on upper troposphere and stratosphere was removed.

In the table σ = P/P_s is used instead of P

Table 1. *Levels of the vertical grid in the NIES TM model*

	H, km	$\sigma = P/P_s$	$\approx\Delta$, m	ζ (σ–θ grid levels), K	Number of levels
Near-surface layer	0-2	1.0–0.795	250	-	8
Free troposphere	2–12	0.795–0.195	1000	- 330, 350	10
Upper troposphere and stratosphere	12–40	0.195–0.003	1000	365, 380, 400, 415, 435, 455, 475, 500	14
			2000	545,	
			–	590, 665, 850, 1325, 1710	
				Total levels:	32

Page 8093, Figure 3: There is something wrong at the poles (kinks).

The latitude of model output data are in a range (- 87.75° – 87.75°), so kinks at the poles are due to wrong interpolation in visualization soft. The figure was replaced with new one.
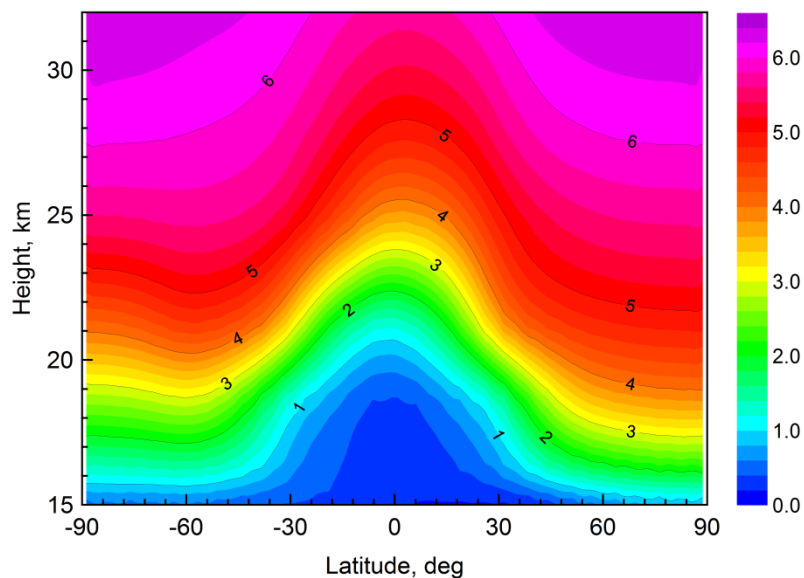
Page 8094: Figure 4 should be given with log(p), however it also might be omitted. Is an average over all stations shown? Clarify in caption, refer at least to text (better repeat sentence in caption).

The caption was replaced with more detailed one:

"Comparison of observed and modelled concentration averaged for the period 2000–2007: a) SF_6, b) CH_4, and c) CO_2. The observed VMRs were derived from six individual profiles of balloon-borne measurements over Sanriku, Japan (39.17°N, 141.83°E)."

Page 8095: Add 'surface volume mixing ratio' in caption. Is this meant here?

Added.

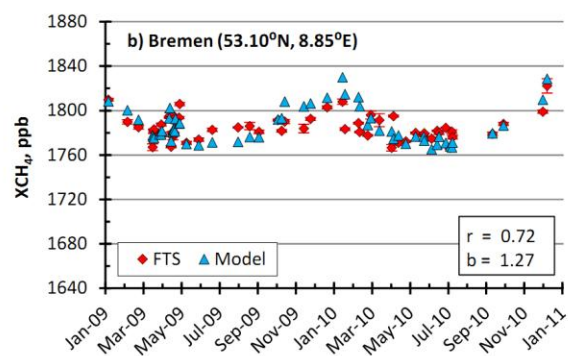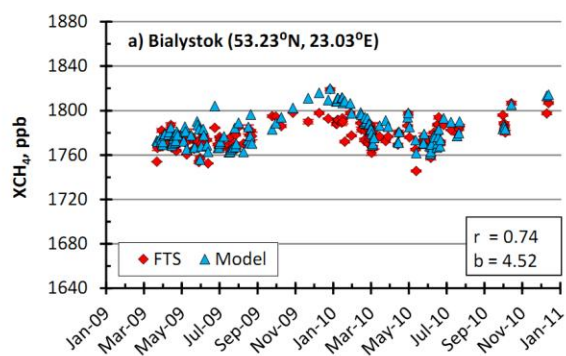**Page 8098: A table would be better here.**

We disagree. A table hasn't any advantage here.

Page 8099: Shown is volume mixing ratio, not concentration, please correct caption.

"concentration" was replaced with "volume mixing ratio".

Page 8101 and 8104: Typos in station names in legends. Please write out XCH_4 and XCO_2 in captions.
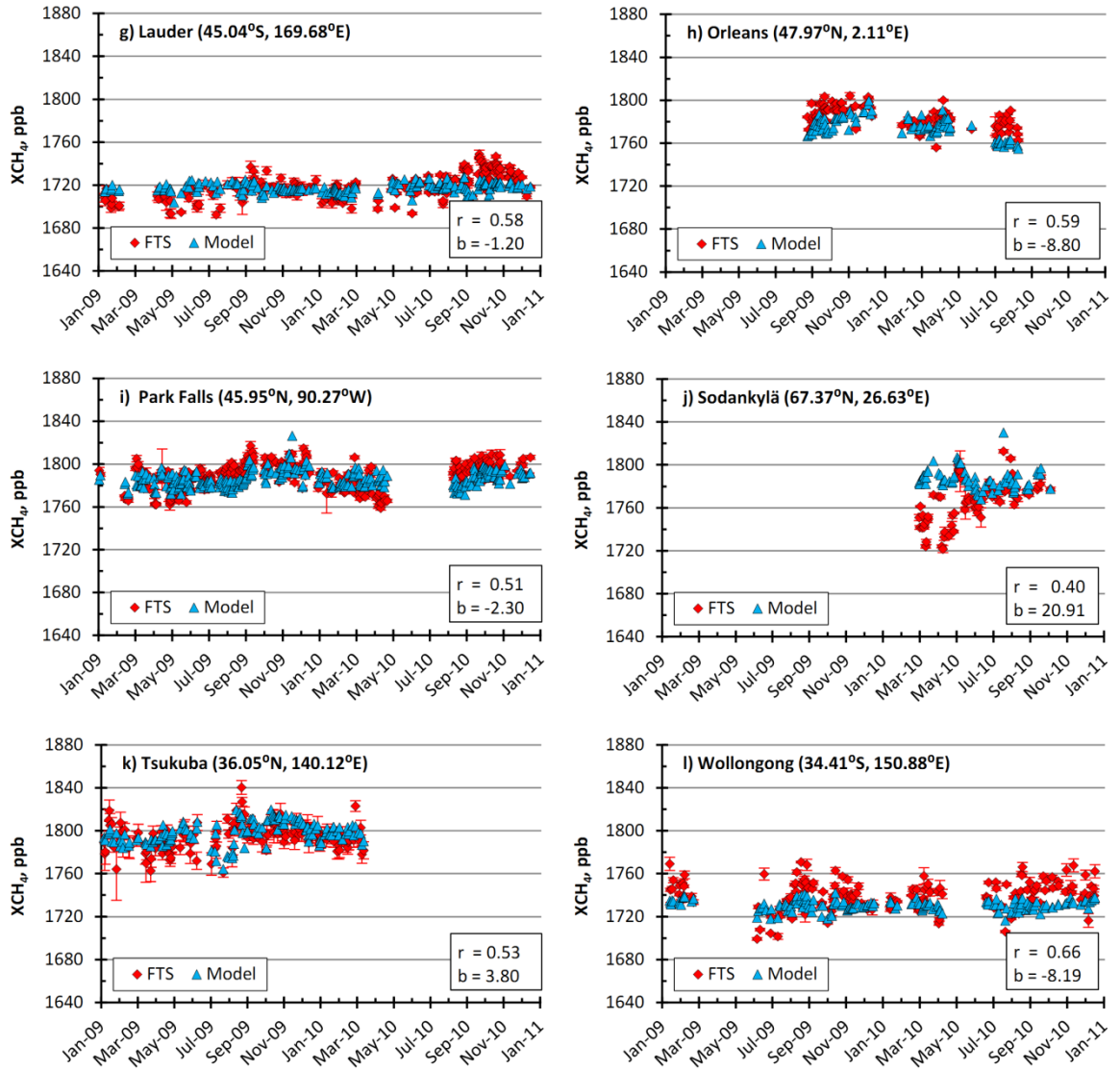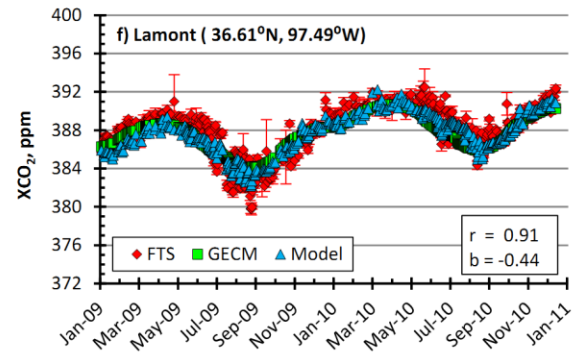
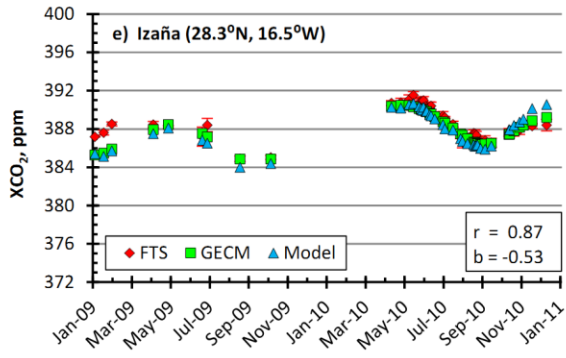Typos were fixed. Captions were corrected.

a) Bialystok (53.23°N, 23.03°E)
r = 0.74
b = 4.52

b) Bremen (53.10°N, 8.85°E)
r = 0.72
b = 1.27

c) Darwin (12.42°S, 130.89°E)
r = 0.55
b = -2.41

d) Garmisch (47.48°N, 11.06°E)
r = 0.44
b = 0.98

e) Izaña (28.3°N, 16.5°W)
r = 0.53
b = 9.05

f) Lamont (36.61°N, 97.49°W)
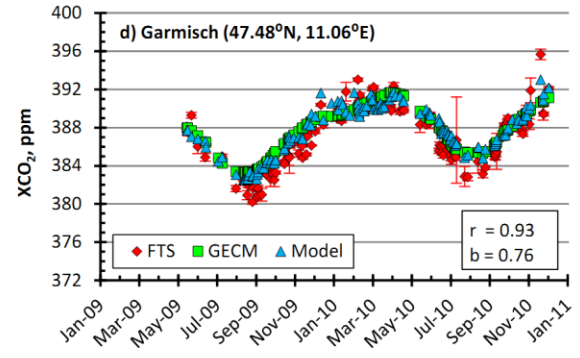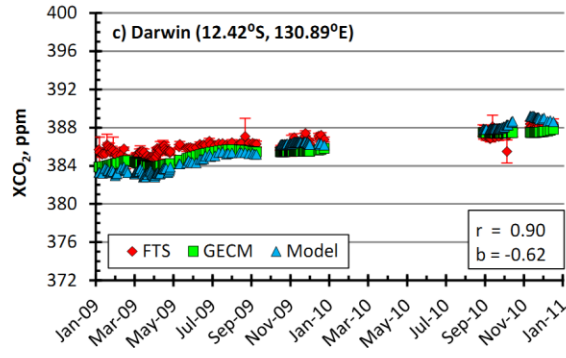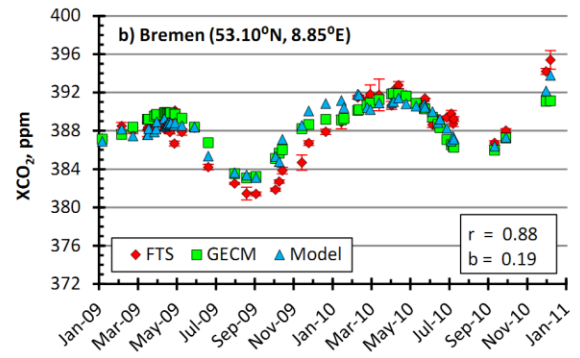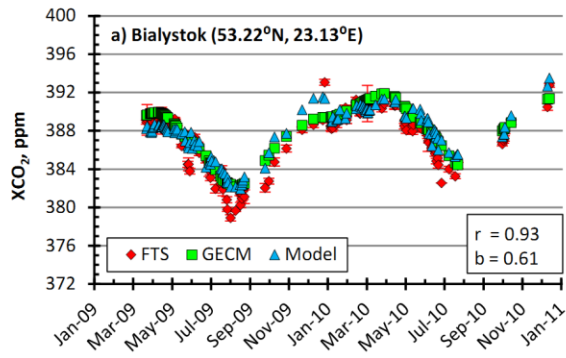r = 0.62
b = -1.13
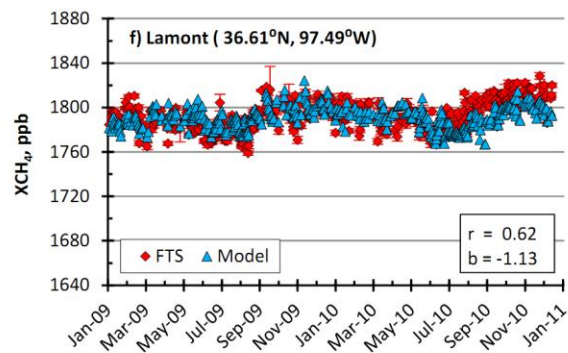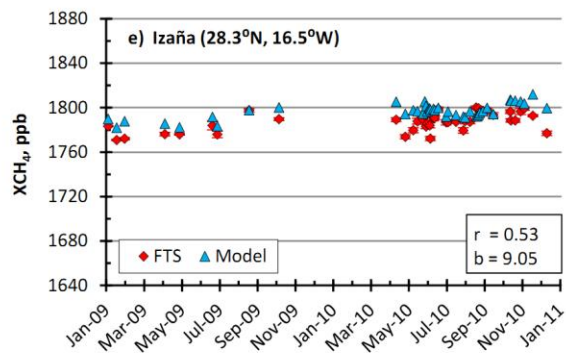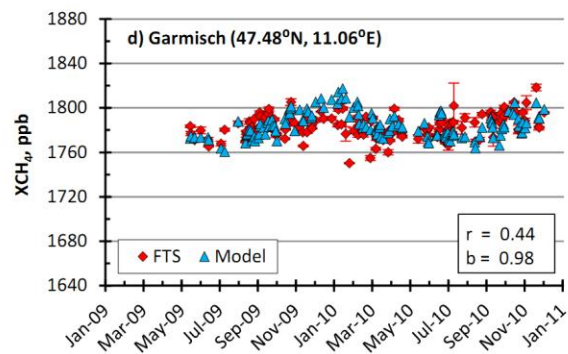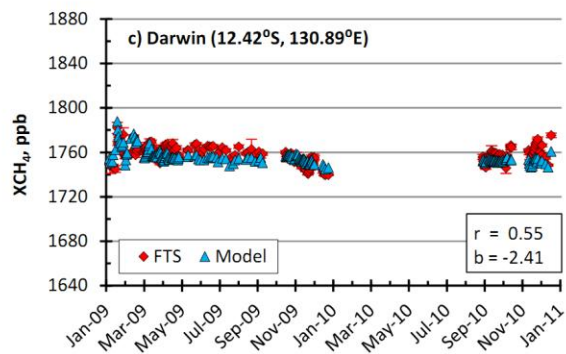
Fig. 11. Time series of XCH$_4$ measured by FTS and modelled by NIES TM for the period January 2009 to February 2011, for the following stations: a) Bialystok (Poland, 53.22°N, 23.13°E); b) Bremen (Germany, 53.10°N, 8.85°E); c) Darwin (Australia, 12.42°S, 130.89°E); d) Garmisch (Germany, 47.48°N, 11.06°E); e) Izaña (Spain, 28.30°N, 16.50°W); f) Lamont (USA, 36.6°N, 97.49°W); g) Lauder (New Zealand, 45.04°S, 169.68°E); h) Orleans (France, 47.97°N, 2.11°E); i) Park Falls (USA, 45.95°N, 90.27°W); j) Sodankylä (Finland, 67.37°N, 26.63°E); k) Tsukuba (Japan, 36.05°N, 140.12°E); and l) Wollongong (Australia, 34.41°S, 150.88°E). The "error" for each symbol is a combination of the spread due to weighted averaging within the 13:00 ± 1 hour local time interval and observation error.

a) Bialystok (53.22°N, 23.13°E)
FTS ◆ GECM ■ Model ▲
r = 0.93
b = 0.61

b) Bremen (53.10°N, 8.85°E)
FTS ◆ GECM ■ Model ▲
r = 0.88
b = 0.19

c) Darwin (12.42°S, 130.89°E)
FTS ◆ GECM ■ Model ▲
r = 0.90
b = -0.62

d) Garmisch (47.48°N, 11.06°E)
FTS ◆ GECM ■ Model ▲
r = 0.93
b = 0.76

e) Izaña (28.3°N, 16.5°W)
FTS ◆ GECM ■ Model ▲
r = 0.87
b = -0.53

f) Lamont (36.61°N, 97.49°W)
FTS ◆ GECM ■ Model ▲
r = 0.91
b = -0.44

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Fig. 12. Time series of XCO$_2$ measured by FTS, modelled by NIES TM and derived from a 3-D CO$_2$ climatology GECM for the period January 2009 to February 2011, for the following stations: a) Bialystok (Poland, 53.22°N, 23.13°E); b) Bremen (Germany, 53.10°N, 8.85°E); c) Darwin (Australia, 12.42°S, 130.89°E); d) Garmisch (Germany, 47.48°N, 11.06°E); e) Izaña (Spain, 28.30°N, 16.50°W); f) Lamont (USA, 36.6°N, 97.49°W); g) Lauder (New Zealand, 45.04°S, 169.68°E); h) Orleans (France, 47.97°N, 2.11°E); i) Park Falls (USA, 45.95°N, 90.27°W); j) Sodankylä (Finland, 67.37°N, 26.63°E); k) Tsukuba (Japan, 36.05°N, 140.12°E); and l) Wollongong (Australia, 34.41°S, 150.88°E). The "error" for each symbol is a combination of the spread due to weighted averaging within the 13:00 ± 1 hour local time interval and observation error.
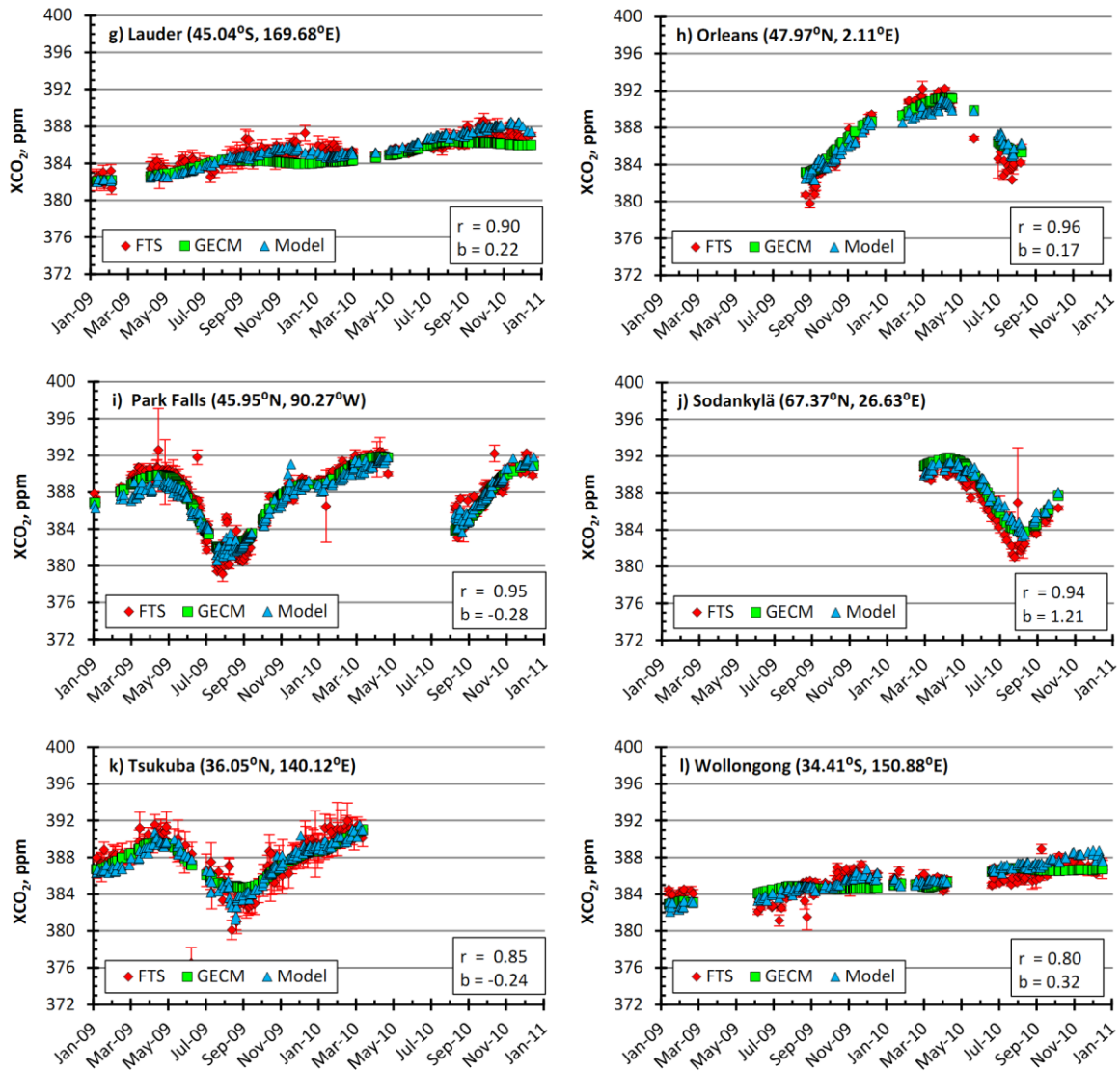
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Page 8103 and 8106: Use more other symbols than circles to allow for better distinction between stations.

We are using different type of symbols for better distinction data from different zones: circles are for stations in the Northern Hemisphere, squares are for stations in the Southern Hemisphere, triangles are for Darwin site, which is located in the tropics. This distribution is very clear for XCH_4 and less visible for XCO_2.

Reference

1. Allan, W., H. Struthers, D. C. Lowe, and S. E. Mikaloff Fletcher, Modeling the effects of methane source changes on the seasonal cycles of methane mixing ratio and δ13C in Southern Hemisphere midlatitudes: J. Geophys. Res., 115, D07301, doi:10.1029/2009JD012924, 2010.

2. Chipperfield, M.P.: New Version of the TOMCAT/SLIMCAT Off-Line Chemical Transport Model: Intercomparison of Stratospheric Tracer Experiments, Q. J. R. Meteorol. Soc., 132, 1179-1203, doi:10.1256/qj.05.51, 2006.

3. Collins, W.D. et al.: Radiative forcing by well-mixed greenhouse gases: Estimates from climate models in the IPCC AR4. J. Geophys. Res., 111, D14317, doi:10.1029/2005JD006713, 2006.

4. Dufour, E., F.-M. Bréon, and P. Peylin: CO2 column averaged mixing ratio from inversion of ground-based solar spectra, J. Geophys. Res., 109, D09304, doi:10.1029/2003JD004469, 2004.

5. Forster, P.M.F., and K.E. Taylor: Climate forcings and climate sensitivities diagnosed from coupled climate model integrations. J. Clim., 19, 6181–6194, 2006.

6. Fung, I., J. John, J. Lerner, E. Matthews, M. Prather, L. P. Steele, and P. J. Fraser: 3-Dimensional model synthesis of the global methane cycle, J. Geophys. Res., 96(D7), 13,033–13,065, doi:10.1029/91JD01247, 1991.

7. Kalnay E.: Atmospheric Modeling, Data Assimilation and Predictability, Cambridge University Press, 364 (ISBN-10: 0521796296, ISBN-13: 978-0521796293), 2002.

8. Lelieveld, J., et al.: Changing concentration, lifetime and climate forcing of atmospheric methane, Tellus, Ser. B, 50, 128–150, doi:10.1034/j.1600-0889.1998.t01-1-00002.x, 1998.

9. O'Brien, D. M., and P. J. Rayner: Global observations of the carbon budget: 2. CO2 column from differential absorption of reflected sunlight in the 1.61 m mbandofCO 2 , J. Geophys. Res. , 107 (D18), 4354, doi:10.1029/2001JD000617, 2002.

10. Oshchepkov S., A. Bril, T. Yokota, I. Morino, Y. Yoshida,T. Matsunaga, D. Belikov, et al.: Effects of atmospheric light scattering on spectroscopic observations of greenhouse gases from space: Validation of PPDF-based CO2 retrievals from GOSAT, J. Geophys. Res., 117, D12305, doi:10.1029/2012JD017505, 2012.

11. Platt, U., et al.: Hemispheric average Cl atom concentration from13C/12C ratios in atmospheric methane, Atmos. Chem. Phys., 4, 2393–2399, doi:10.5194/acp-4-2393-2004, 2004.

12. Saito, R., Patra, P. K., Deutscher, N., Wunch, D., Ishijima, K., Sherlock, V., Blumenstock, T., Dohe, S., Griffith, D., Hase, F., Heikkinen, P., Kyrö, E., Macatangay, R., Mendonca, J., Messerschmidt, J., Morino, I., Notholt, J., Rettinger, M., Strong, K., Sussmann, R., and

Warneke, T.: Latitude-time variations of atmospheric column-average dry air mole fractions of CO2, CH4 and N2O, Atmos. Chem. Phys. Discuss., 12, 5679-5704, doi:10.5194/acpd-12-5679-2012, 2012.