Reply to Anonymous Referee #1

Prabir Patra and his colleagues from the TransCom modeling community present here a comprehensive description of the major results and new findings from the global TransCom methane model intercomparison exercise. The manuscript is very interesting to read, also for experimentalists and well suited for publication in ACP. I am particularly pleased that the results from these model simulations will be made available, and hopefully easily accessible also for the measurement community that is not so experienced in handling Gigabytes of global model output data that needs special software to be digested. (At this moment, however, I have not been successful to access the simulation results via the links reported in section 4 of the manuscript…)

Thank you very much for reading the article and providing constructive comments. Please find below our replies (text in black) to your comments (text in grey).

We have considered making the TransCom-CH4 model output available from WDCGG, which currently does not seem a viable option. In any case the data will be made available upon request (e-mail: prabir@jamstec.go.jp). Furthermore, we are thinking of putting the model time series at selected sites at EBCRPA data server, and the link will be modified accordingly (http://ebcrpa.jamstec.go.jp/dav/prabir/transcom-ch4/sites_data/).

Besides a number of minor suggestions for changes or clarifications in the text and figures, I have one major point, which needs to be addressed before publication of this manuscript in ACP. This concerns the interpretation of the observed change of SF6 Inter Hemispheric Gradient (IHG) in terms of a change in the inter-hemispheric exchange time (IHET) of air masses. Patra et al. use their Equation 4 to estimate this exchange time from measurements and also from modeling results of hemispheric mean concentrations. But in both cases, besides the concentrations and their temporal changes, calculating the IHET also requires to know the ratio of emissions in the northern and the southern hemisphere i.e. En/Es. In fact, a trend in "observed" and "modeled" exchange time, as found in the present study, may also show up if a change of the distribution of emissions was not properly taken into account in the emissions ratios used for the period in question. Although it is straightforward, and potentially very accurate, to estimate total global annual SF6 emissions only from observed mean growth rates of SF6 in the atmosphere, correctly determining its distribution is a very difficult task. It is not surprising that inter-hemispheric exchange times derived from observations and from model simulations agree well when the model results properly reproduce the observations at the respective background stations used for this estimate. However, this does not automatically imply that the trends in the IHET are correct. As has been discussed e.g. by Levin et al. (2010, cited in the manuscript), a tracer with a completely different distribution of emissions (than SF6) could help to confirm (or falsify) the estimated changes of the IHET.

We actually have tested sensitivity of IHET results to the emission ratio, which did not show any significant influence on the calculated exchange time. Figure R1 (below) shows the comparison of using variable emission ratio (ER) = E_n/E_s vs. using a constant ER of 30.73. The final outcome (top panel) did not change appreciably, which explains why these results were not included in the manuscript. We have added the following sentence to the revised version (section 2.5) "We have tested that the estimated τ_{ex} does not depend strongly on the E_n/E_s ratio (ref. reply to reviewer#1)." An additional test has been conducted for forcing a constant τ_{ex} by changing E_n/E_s ratio, which suggests dramatic decrease (from 24.7 to 8.7) in E_n/E_s is required for keeping the IH exchange time near constant during 1996 and 2002. A plausible reason for such a decrease in E_n/E_s ratio cannot be established in this study.

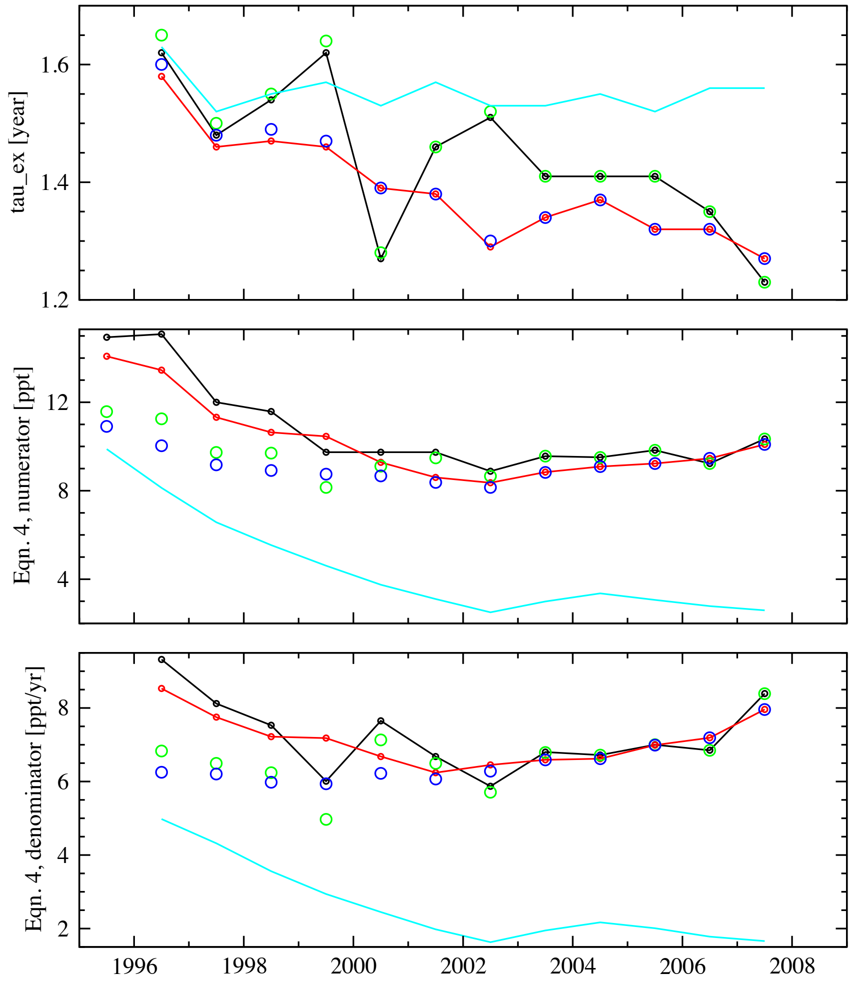

Figure R1: Comparison of estimated τ_{ex} (top panel), and the numerator ($\left[\Delta c_{n-s}\left(\frac{E_n}{E_s}+1\right)\right]$; middle panel) and the denominator ($\left[\frac{E_n}{E_s}\frac{dc_s}{dt}-\frac{dc_n}{dt}\right]$; bottom panel) in Eq 4. The original calculations are show as solid line for observations (in black) and ACTM simulation (red), while those using constant ER value of 30.73 are shown in green and blue circles, respectively. One case is prepared by changing the E_n/E_s ratio to derive a constant τ_{ex} values (cyan line), where E_n/E_s = 30.7, 27.7, 24.7, 21.7, 18.7, 15.7, 12.7, 10.7, 8.7, 9.7, 10.7, 9.5, 8.3 and 7.1 are used for the years 1994-2007, respectively.

Further comments:

Abstract:

It would be helpful if a few words about the meteorological fields (i.e. that they are different in the different models) were mentioned.
Mentioned "All but one model transports are driven by reanalysis products from 3 different meteorological agencies."

Line 44: The term "CH4 tracers" is a bit confusing for the non expert. Better use e.g. "scenarios" (throughout the text).
We have chosen the term 'scenario' for CH4 tracers as suggested.

Line 56: See major comment above. What is calculated here is not an exchange "rate" but an "exchange time" or "hemispheric residence time". This term needs to be corrected throughout the manuscript.
Good suggestion – we have modified "exchange rate" to "exchange time"

Line 63: I guess what is meant is not CH4 "budget" but CH4 "distribution". The term "budget" is confusing here as it is normally used for a total inventory.
Here we mean to say "budget" as it is discussed later on how the STE rate, inferred from vertical gradients in the stratosphere effects the CH4 lifetime.

Line 67: The term "as noted by the 1sigma" is confusing; I would just leave that out. Is it so surprising that the life times do not vary much when the OH fields and reaction rates are pre-set? Generally, I think that, although there were no real scenarios of the sink processes tested in this study, the influence of the sink processes on the CH4 and MFC distributions should be more thoroughly at least discussed.
We did extend the discussion with respect to uncertainties in the OH distribution (IH gradient) and how they may affect the CH4 simulations, as per the comments by review#2 and Dr. James Wang.
Following your suggestion "as noted…" part of this sentence is deleted.

Introduction:

Line 75: Unfortunately, the WDCGG data base still does not give a "complete" list of observational programs.
We agree, but the most comprehensive list of the measurements sites and organizations worldwide are available from WDCGG (along with the well calibrated data).

Line 93: May be it is worth to mention here that these are now bottom-up estimates of individual flux components.
Mentioned

Line 99: Explain CTM (and ACTM, line 185, at least once)
CTM is now defined in the Abstract, and ACTM is expanded here.

Line 117: I have not seen any discussion on diurnal cycles in this manuscript.
We have added a sentence here "Only the first two, IH gradient and seasonal cycles, are discussed in this paper."

Line 139ff: This sentence needs re-formulation: Why should e.g. inert tracer transport of SF6 have implications on chemically active species?
Reformulated as “transport on the lifetimes and distributions of chemically active species (CH_3CCl_3, CH_4),…”

Section 2:

Line 163: I assume the reaction rates have units, e.g. 1/s.
Yes, the units are cm^3 $molecule^{-1}$ s^{-1}. Added

Line 184: Add “(MFC)” after CH3CCl3.
MCF is added

Line 192: What about tropospheric Cl as a sink for CH4? (possibly of similar size as the soil sink, see Allan et al., JGR 112, D04306, 2007)
We do not have a good grasp on tropospheric Cl distribution to incorporate in such a model inter-comparison study. However, please note that the budget of CH4 emission, sinks and atmospheric growth rate will be disturbed significantly from that is presented here, if an additional sink mechanism is to be incorporated. Such sink mechanisms have to be tested in single chemistry-transport model frameworks first. For example, we do not have access to a 3-dimensions distribution of Cl radicals in the troposphere (like that of tropospheric/stratospheric OH, stratospheric Cl & O1D).

Section 2.2 and Table 1: The explanations in Table 1 are a bit brief. This makes it necessary, when looking at the results, to always go back to Sec. 2.2 for more explanation. Also, it is not really clear to me which anthropogenic emissions were used in scenarios (“CH4 tracers”) 3-6, i.e. EDGAR 3 or EDGAR 4?
Table 1 is modified to indicate the use of Natural and Anthropogenic emissions for CH4.

Lines 281ff: Has a seasonal cycle of 222Radon exhalation rate been used in this study (as is generally measured at sites with fine grained soils)? The potential influence of seasonal 222Radon emissions will have to be kept in mind when comparing the simulated results with observations.
We do not compare the Radon simulations with observations, but 2-3 dimensional distributions are used to characterize transport model differences in relation with deep cumulus convection, which does not rely on any assumption regarding the seasonality of the emissions.

Section 2.3, lines 306/307: I do not understand this sentence “… and do not automatically …”
Sometimes it is assumed that one meteorological reanalysis or the higher spatial resolution CTMs are better in simulating chemical species. However, that is not the case – this is just a warning before reading the rest of the paper.

Section 2.4, line 375: Please give a reference for “Pearson’s moment correlation analysis”
We have given reference to Numerical Recipe (Press et al., 1986), which is what used for the correlation calculations here.

Section 3:

Line 410: I guess it should read "Figs. S1-S17".
Yes – done.

Line 427: Include a "more", i.e. … more "leaky"
Done

Line 448: I guess it should be "Figs. S2 – S4".
Changed to "Figs. S2 and S4"

Line 470ff: As mentioned earlier, estimating the global SF6 emissions trends should not be model-dependent. One could even think of estimating this simply from observed trends at one representative site in the northern and one in the southern hemisphere (i.e. without any model).
Yes, we agree for SF6. This is kind of a prelude to what we is discussed for CH3CCl3 (intermediate complexity) and CH4 (difficult to model) later on.

Line 486ff: "These lifetimes … (Prinn et al., 2005)" This sentence is unclear to me.
Modified as "Although these lifetimes in the stratosphere and troposphere are within the range of independent estimates, …yr, respectively (Prinn et al., 2005), both the mean values are lower for ACTM."

Line 498: I would say that STE uncertainty should not be tracer-dependent. The EFFECT of STE on the tracer DISTRIBUTION may be different (for CH4 and CH3CCl3).
Good point. Modified as "…surface emissions and the effect of STE are less certain than for…"

Line 509: I would like to see a bit more discussion on the point made here: This is why also SF6 and 222Radon have been modeled (to check transport differences between the models), right?
Some of these dependences are addressed in details later. We modified the end of this sentence as "…also play a role (details in sections 3.3 and 3.4)"

Line 545: "… noting that the OH loss is realistically represented, as seen in the CH3CCl3 seasonal cycles." I do not totally agree to this statement, e.g. there is (1) a significant phase shift in the MFC seasonal cycle between models and observations, and (2) noting that CH4 in the southern hemisphere troposphere is mainly sink driven, the seasonal amplitude of this sink seems to be over-estimated by all models by about 20%.
We agree that the simulated CH4_CTL seasonal cycle amplitudes overestimate the observations and that there are phase differences in CH3CCl3 seasonal cycles at CGO. On the other hand, the simulated CH4_CTL seasonal cycle phase and CH3CCl3 seasonal cycle amplitude are in very good agreement with the observations. If we move to more remote sites such as South Pole there is no apparent disagreement between the observed and simulated CH4_CTL seasonal cycles (Fig. R1). Unfortunately, no clear statement can be made for the phase shift in CH3CCl3 seasonality at South Pole. Figure R1 also shows that transport differences are larger

than the differences arising from the use of other OH distributions in ACTM and GEOS-Chem.

Note also that the agreement between modeled and observed seasonal cycles varies significantly for both CH4_CTL and CH3CCl3, which is clear from Fig. R1 (because of a larger number of years). The amplitude and phase of the observed cycles do change from year to year, which is not well captured by the models. However, we cannot address this issue in this paper, since we do not represent inter-annual variability in OH.

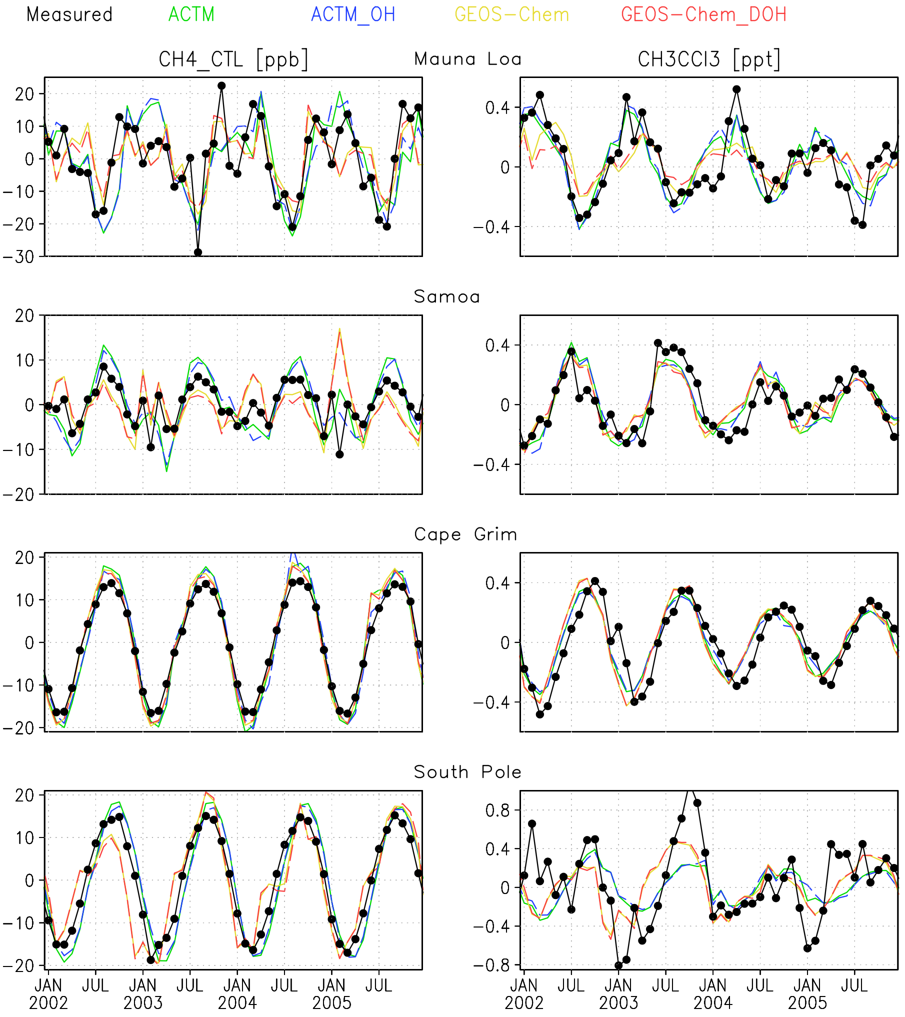

Figure R2: Same as Fig. 5, but seasonal cycles of CH4_CTL and CH3CCl3 are only shown for a longer period (4 years) and 4 sites. Only 2 model results having different OH distributions are shown here.

The overestimation of CH4 seasonal cycle at CGO or SPO does not exist for the CH4_EXTRA scenario (Fig. R2a).

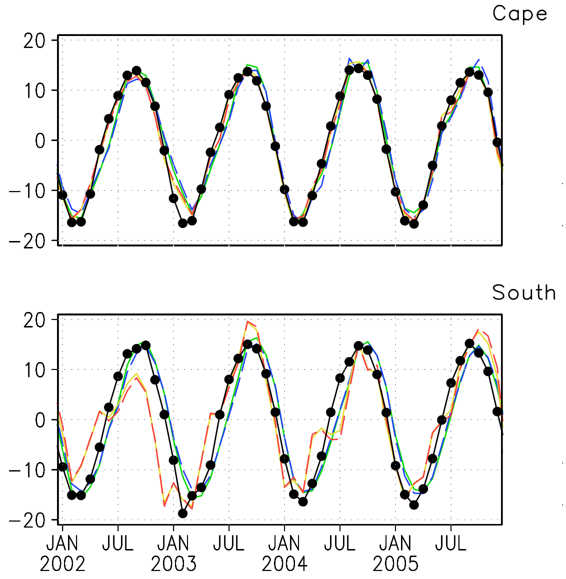

Figure R2a: As in Fig. R2, but the CH4 emission scenario of EXTRA (CGO: top panel; SPO: bottom panel).

Line 630: I would replace the word “differences” by “range”
Modified.

Line 632ff: As mentioned above, it is a bit misleading to talk of “observation-based” vs. “model-based” inter-hemispheric exchange times. (But this whole paragraph needs to be revised as explained earlier…)
This sentence is modified as:
The average tau_ex of 1.39 yr is in excellent agreement with the estimates of 1.3 yr (Geller et~al., 1997) and 1.5 yr (Levin and Hesshaimer, 1996), derived using measured SF6 time series.

Line 654: I cannot find such a Figure S17.
Changed this to “Fig. 9”.

Lines 677ff: I think this interpretation (of constant vertical gradients) needs to be explained in a bit more detail.
This is because the averaged CH4 vertical profile gradient in the stratosphere is dominated by the CH4 loss due to the reactions with Cl, O^1D, OH, and model transport property. This reiterates the fact that the models spin-up were completed before 1992, and that the stratosphere-troposphere exchange (STE) rates were well established for individual model depending on their own behaviour. (which is what is stated in the next sentence).

Line 688: Perhaps include “(Eq. 5)” after “…at 8 sites.”
Included

Lines 702ff: Why should the OH estimates from MFC become less uncertain after 2000? May be the method becomes less sensitive because MFC is strongly decreasing? Or is it that the variability of MFC is simply getting smaller? I cannot

find the citation Montzka et al., 2011, in the references.
Because the errors in emission become less relevant as the photochemical loss exceeds the emission. Montzka et al. reference is added to the bibliography.

Conclusions:

Line 766: What is meant with "suppressing the seasonality of the underlying emissions"?
This word however is changed to 'concealing' as suggested by review#2. We have added following text to make the meaning clearer.
"The simulated seasonal cycles of CH4 are influenced by the photochemical destruction by OH: without destruction, the simulated seasonal cycles would more strongly reflect the seasonal cycles present in the emission scenarios."

Line 769: I guess it should read "A set of six global CH4 flux representations ..."
The word 'global' is removed.

Line 801 – end: This sentence needs to be re-formulated.
Modified as :
We find net CH4 emissions to the atmosphere of 513±9 and 514±14 Tg-CH4 year-1 (soil sink subtracted) for the 1990s and 2000s, respectively, are consistent with the atmospheric losses accounted for due to OH, O1D and Cl in order to simulate CH4 concentrations and growth rates at the surface sites.

Figure 1: It is a bit confusing that the surface/soil sink shows up in two boxes, dark green and pink.
We have removed 'soil sink' term from the green box.

Figure 2: I cannot find the black (CTL) line in Figure 2a.
The black line is hiding behind the red line because the seasonal cycles of CTL and CTL_E4 are identical. A note in the figure caption is added.

Figure 4: It would be interesting to also see the starting values of SF6 in the different models. Please note in the caption how the "difference" is defined, i.e. observed – simulated or vice versa. The averaging interval for the troposphere (1000-200mbar) is not consistent with the caption in Fig. S19 (850-200 mbar).
Simulated SF6 concentrations are available from 01 Jan 1990 onwards. Unfortunately we did not archive the spin up period (1988 & 1989), which would have allowed us to verify the applied initial condition.

Figure 7: Please note in the caption which CH4 scenario is displayed here. It is interesting to note that all models show a steeper IHG for CH4 than observed. May be this is due to some bias in the anthropogenic CH4 emissions distribution (compare remark on inter-hemispheric exchange times).
Changed from CH4 to CH4_CTL to clarify the scenario.
We agree that the model IHGs are a bit stronger, by ~20ppb, than that observed for CH4 in the early part of the simulation (until ~2000), but the model results seem to be spreading across the observed IHG line fairly uniformly for the post-2003 period. While the CH4_VISIT scenario shows similar behaviour, the CH4_E4 scenario shows consistently overestimated IHGs by the models (Fig. R2). It is also possible that the

OH amount should be a bit higher in the northern hemisphere (NH). The simulated IHGs for CH3CCl3 are a bit overestimated too for most of the years.

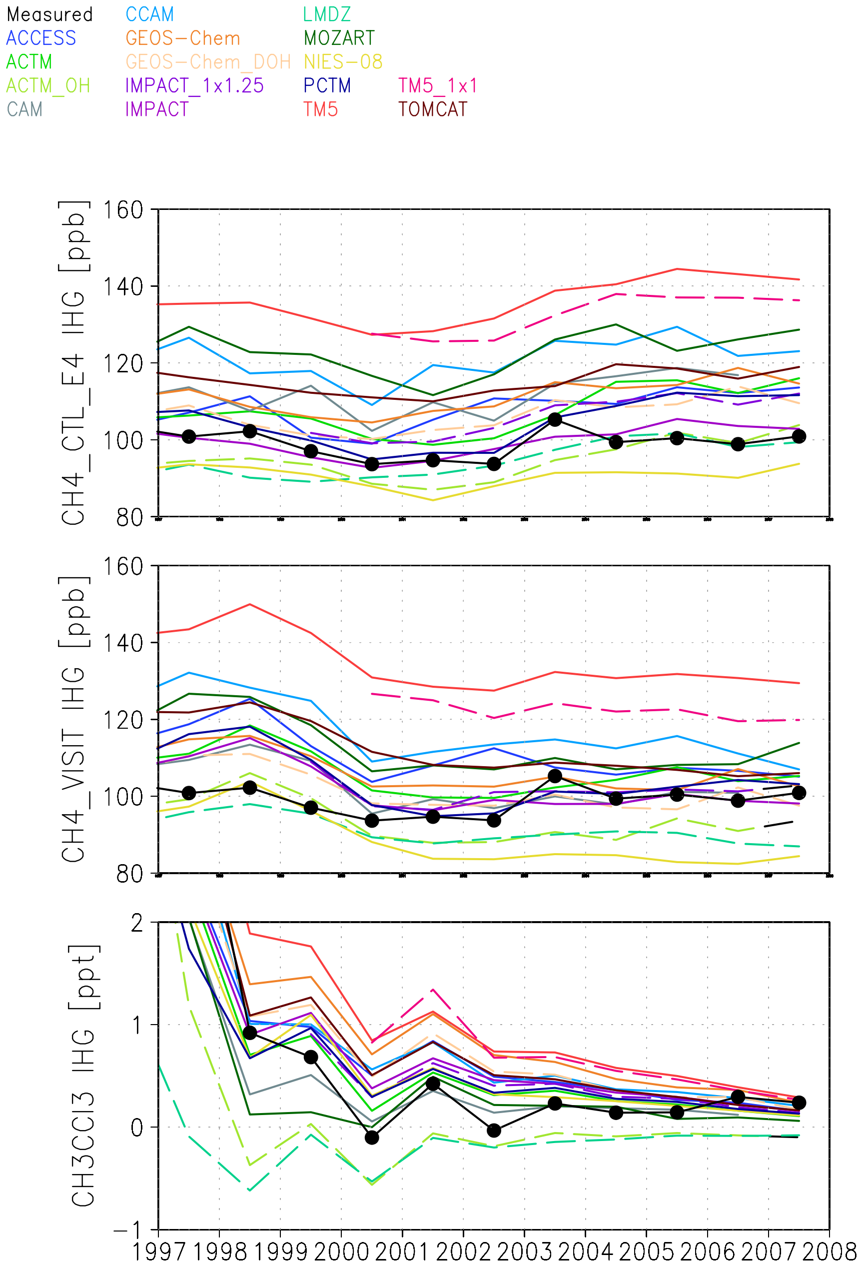

Figure R3: Comparisons of simulated and model IHGs for two CH4 emission scenarios (top two panels) and CH3CCl3 (bottom).