Reply to Anonymous Referee #2

GENERAL COMMENTS
This manuscript describes the findings from the recent TransCom model intercomparison effort, and specifically the analysis of the role of emissions, transport and chemical processing in driving global CH4 distribution and changes. The methodology followed is sound and the results of the analysis are interesting and useful for future discussions on CH4-related topics. The manuscript is certainly well suited for publication on Atmospheric Chemistry and Physics, although I would like to see the following general changes being made, as I believe they will improve the manuscript significantly:

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).

First, I feel that the discussion on OH is, in general, more limited than it should be. In particular, there is not as much discussion as I would expect regarding the runs that used an alternative OH distribution (e.g. how different the OH distribution is and why; how these differences link to features of the simulated CH4 and CH3CCl3), and there is not any discussion as to how the results might differ if the assumed OH distributions were different.

To discuss the implications of IHGs in Fig. 9 for NH/SH ratio of OH abundance, we have added this text:
Two models submitted simulations using different OH, which can be used for understanding the role of IH gradients in OH on the simulated CH4 and CH3CCl3 IH gradients. The NH/SH ratio of hemispheric total tropospheric OH are (1) 1.32 for ACTM OH and (2) 1.11 for GEOS-Chem DOH, while that for TCOM OH (Spivakovsky et al., 2000) is 0.99. Now the observed CH4 IH gradient is 96 ppb, and we get ACTM and ACTM OH simulated IH gradients as 100 and 88 ppb, respectively, Which means the NH/SH OH ratio should be 1.1 [= (1.32 − 0.99) × (100 − 96)/(100 − 88)] for simulating the observed CH4 IH gradient by ACTM. This is similar to the NH/SH OH ratio of GEOS-Chem DOH, and CH4 IH gradient simulated by GEOS-Chem DOH matches well with the observations. However, GEOS-Chem DOH does not simulate well the IH gradient for CH3CCl3, a species that has been used for benchmarking tropospheric OH concentrations and distributions. Given the small amount of alternative OH distributions and models and the remaining uncertainties in CH4 and CH3CCl3 emissions, our best judgent at the moment is that we cannot falsify the NH/SH gradient (0.99) derived by Spivakovsky et. al. (2000).

A focused study in the future will help to diagnose the NH/SH ratio of OH more accurately.

Furthermore, there are some parts of the manuscript where the messages are not clear, due to the sentences being too long and/or confusing. I include comments below which I believe will improve this aspect of the manuscript, and will eliminate some typos and minor issues that I encountered.

SPECIFIC COMMENTS

We have accounted for all the specific comments during revising the ACPD paper. Thank you very much for these help.

Abstract

You mention that up to 60% of the variability is explained by variations in biomass burning and wetland emissions. The reader does not encounter a part of the analysis clearly demonstrating that.
We reordered these two sentences with modifications as follows:
"Using six sets of emission scenarios, we show that the decadal average CH_4 growth rate likely reached equilibrium in the early 2000s due to the flattening of anthropogenic emission growth since the late 1990s. Up to 60% of the interannual variations in observed CH4 concentrations can be explained by accounting for the interannual variations in emissions, from biomass burning and wetlands, as well as meteorology in the forward models."

Section 1 Page 18770, Line 15: Please change "past few years" to "past two decades".
Done

Page 18770, Line 24: Please change yr1 to yr^{-1}.
Done

Page 18771, Lines 14-17: The reference to Shindell et al. (2009) is important, but this sentence is confusing as it does not exactly reflect the findings of this study (26% is the reduction of OH due to CH4 emissions, while 40% is the change in CH4 GWP due to the inclusion of oxidant-aerosol interactions). Please rephrase.
Changed to "According to Shindell et al. (2009) the 100-yr integrated GWP of CH4 is sensitive to changes in oxidant-aerosol precursor emissions and to OH-feedbacks of CH4 emissions itself. Proper understanding of the CH4 budget is crucial for these assessments."

Page 18772, Line 13: Please add "," before "on CH4". Section 2
Done

Page 18772, Line 22: Referring to the model as "Chemical-transport" rather than "Transport" sounds more accurate. Perhaps it should be changed throughout the text.
Done.
The way we understand a CTM is a model including a full photochemistry scheme, while our simulations make use of transport and sources and sinks of the target tracers only. Thus we haven't made similar changes at other places.

Fig. 1: It would make more sense to write "CH4 emission" rather than "CH4 flux" in the green box, since "flux" can include other aspects (e.g. chemistry), and since "emission" is used as a term for CH3CCl3, Radon and SF6 in this figure anyway. Also, I am not sure if "soil sink" should be included in this box, since it is already in the pink box, on the right.
Thank you. Done

Page 18773, Lines 14-16: It will not be clear to all the readers how these stratospheric processes are treated. In particular, where do O^1D and Cl concentrations come from?

Are they tabulated numbers coming from the Cambridge 2D model, which are then read-in and multiplied with the k values of the reactions? Also, maybe OH was not meant to be in line 14 (as you have already explained above how OH is handled)?
We have not incorporated the O^1D and Cl concentrations explicitly in this experiment, but the parameterized loss rates (reaction rates x concentrations).
OH is removed from this sentence.

Page 18774, Line 1: Please specify that NH and SH stand for northern hemisphere and southern hemisphere, since it is the first time that they are being used.
Done

Page 18774, Line 7: Please add "and interpolated on each model's grid" after "ACTM (Patra et al., 2009a)", in order for the reader to understand what "interpolated" mean in line 12. Also, please add a full stop before "Because".
Done.

Page 18774, Lines 13-15: Please remove parenthesis from the sentence starting with "Similarly", since this statement is fairly important.
Done

Page 18774, Line 17: Please rephrase to ". . .are provided by Krol et al.". . .
Done. Modified as "…are provided by Krol et~al. (1998; see also Kanakidou et~al., 1999)."

Fig. 2: Apparently the authors have chosen 1997-99 as an example period to demonstrate seasonality in emissions. It would be good to state this (i.e. that this is an example period) or, alternatively, show and average seasonal cycle from all years. Also, please change "flux" to "emission" in the figure caption, since "flux" could also imply other aspects (e.g. chemistry-related). Finally, note that there is no black line in this figure.
This part of the caption is changed to "Examples of CH4 emission seasonalities corresponding to the years 1997 and 1998 are shown in (a) (the black line is hidden behind the red line), and annual mean CH4 emissions for the period of 1988--2008 are depicted in (b)."

Page 18775, Line 15: Please change the sentence to read like "The following source and sink components of CH4 were considered in the six different experiments listed in Table 1:"
Modified as "...the six different scenarios listed…"
We have not used the word 'experiment' because we are calling this whole exercise as TransCom-CH4 experiment

Table 1: I do not agree with the term "tracers" being used here. It would be clearer if you name them "experiments" or "scenarios", not just here, but throughout the manuscript. Also, Table 1 is central to the paper, but the "DESCRIPTION" column is a bit hard to follow. I would suggest that the abbreviated terms that are being used should be explained in the caption. Some of them may have already been explained in Section 2.2, but the descriptions are scattered and not easy to pick from there.
We agree with you and the other reviewer for replacing 'tracer' by 'scenario' throughout the manuscript. However, we use the term tracer for referring to all these

species in a general sense – a sentence is added in the last paragraph of the Introduction as “Simulations of SF6, 222Rn, CH3CCl3 and six CH4 emission scenarios are commonly referred to as model tracers.”

Page 18775, Line 16: Please change to “. . .(IAV ANT), based on. . .”, since the way it is, it may seem to the reader that these emissions are used for all the experiments.
Done

Page 18775, Line 21: Please change to “Anthropogenic emissions (IAV ANT E4), based on a more advanced EDGAR. . .”
Done

Page 18775, Line 26: Similarly to above, please change to “and termites, based on. . .”
Done

Page 18776, Line 2: Please remove “the” and “are”. Page 18776, Line 25: Please remove “that”. Page 18776, Line 26: “CYC BB” has not been defined.
All done, and line 26 is modified as “Thus biomass burning emissions in CYC NAT is only partially…”

Page 18777, Line 21: Please delete “to be”.
Done

Page 18778, Line 12: Please separate “variedfrom”.
Done

Page 18778, Line 15: Please change “linked” to “link”.
Done

Page 18779, Lines 3-6: Please split and rephrase this sentence, as it is a bit too long and hard to follow.
Modified as “The TransCom-CH4 experiment archived model simulations for 18 yrs and 9 tracers. We have sampled model output at 280 surface sites and 115 vertical profile sites (at all model levels within troposphere) at hourly time intervals. 3-D monthly mean mixing ratios at 17 standard pressure levels for 1990--2007, and noon-time daily values for 2001--2007 are also archived.”

Page 18780, Line 21: Please change “model” to “modeled”.
Changed to ‘simulated’

Page 18781, Line 13-15: I suggest that the contents of the parenthesis are made a separate sentence.
Modified as “…, respectively. Ideally vertical distribution properties of each species should be accounted for with appropriate air mass factors for calculating B.”

Section 3

Fig.3: Please indicate the units of pressure on the left side of the actual figure. Also, please indicate in the caption to which model run these results correspond.
Y-axis is labeled

Page 18782, Line 16: Please add “more” before “leaky”.
Done

Page 18783, Line 22: Please add “(not shown)” after “sites”.
Done

Fig. 4: Please change caption to read like “. . .annual mean CH4 in CH4_CTL. . .” (since, as I have suggested, CH4_CTL should not be referred to as a “tracer”. Also please replace “,” with “:” before “MLO”.

Done

Page 18783, Lines 25-28: The sentence does not read easily and needs to be rephrased. Also: do you have a feeling as to why ACCENT and ACTM_OH have this distinct behavior after 2000? Finally, it is not clear how this sentence links to the one after it.
Modified as “Only ACCESS shows increasing differences in time until 2006. ACTM_OH, which uses EDGAR4.0 emissions without scaling between 1988--2005, and 2005 emission for 2006 and 2007 produces slower increasing concentrations compared to observations after 2000.”
The 2nd sentence here is linked to the one after it. Hope this is better. We do not have clear ideas regarding the ACCESS behavior.

Page 18784, Lines 8-13: This is a long and not clear sentence. Please spit it in two and delete the first “Because”.
Modified as:
The lifetime of CH3CCl3 due to photochemical removal is much longer in the stratosphere (~28.6 yr) than in the troposphere (~5.8 yr) (estimates from ACTM simulated loss rates at model grids). The troposphere to stratosphere transport plays a minor role in the global total budget of CH3CCl3 after the late 1990s, because the concentration gradients across the tropopause reduced to less than 10 ppt (ref. Table 3).

Page 18784, Line 13: Please rephrase sentence starting “These lifetimes. . .”.
Modified as “Although these lifetimes in the stratosphere and troposphere are within the range of independent estimates, …, respectively (Prinn et~al., 2005), both values are lower for ACTM.”

Page 18784, Paragraph starting at Line 13: Although CH4 is the focus of this study, this paragraph discusses CH4 too briefly, compared to the previous paragraphs. I would like to see some more discussion and perhaps some strengthening of the conclusion that transport differences are the ones causing the features mentioned.
The transport and chemistry differences between the models and their effect on CH4 and other species are discussed later in detail. Kindly refer to the dedicated subsections 3.3 and 3.4.

Figure 6: Please put “Flux: CTL” and “Flux: EXTRA” at the top of the panels and change flux to “Experiment” or “Scenario”. Change caption accordingly.
Done

Page 18787, Line 26: CH4_CTL_E4 seems to consistently have the least good performance in terms of IAV. This is worth mentioning and commenting on in the text.
We agree – please note that this has been mentioned in the next paragraph

Page 18788, Line 12: "Their estimate is largely inconsistent with our results. . .": I am not sure where is this inferred from.
We mean to say that Lamarque et al. suggested that a decrease in CH4 emissions is needed to explain the observed steady state in CH4 in the early 2000s. However, our simulations reach this CH4 steady state without decreasing emissions.

Page 18788, Lines 19-20: This statement is not that well supported by the previous sentences. Why is it emissions and not e.g. transport or temperatures that are causing the problem?
We think that the EDGAR4.0 emissions are increasing too fast in the period 2001-2005 because it is a consistent feature of all our CTMs, which includes interannual variabilities in transport and temperature.

Page 18789, Lines 1-2 (and last lines of previous page): Do we understand what is the main contributor to this reduction in uncertainty. Please comment.
One of the main reasons could be that all (but ACCESS) models are driven by reanalyzed meteorological data (U, V, T etc.) as opposed to Denning et al.

Page 18789, Line 2: Please change "is" to "are" and add a "to" after "compared".
Done

Page 18789, Line 9: "This behavior. . .": Obviously this statement refers to the over-all trend of increasing IH exchange rates and not to the IAV observed in the periods mentioned (1996-99, 2004-07). Please make clearer.
Changed to "This decrease in tau_ex…"

Page 18790, Line 16: Please remove "but".
Done

Page 18790, Line 21: Please change "is" to "are".
Done

Page 18790, Lines 20-22: I believe that "vertical mixing" may be a better term than "troposphere to stratosphere transport". This is because the main point is not that CH4 is transported solely to the stratosphere, but rather to higher altitudes in general (either tropospheric or stratospheric), where it progressively undergoes less destruction.
Actually the turnover time of airmass in the troposphere is much shorter than in the stratosphere, e.g., the air in the lower troposphere can move to the upper troposphere and vice versa within days/weeks, while the turnover time between the lower and upper stratosphere can be up to several years (e.g., Patra et al., ACP, 2009). As a consequence, the simulated exchange time between the troposphere and the stratosphere can significantly influence the global CH4 lifetime and growth rates simulated at surface sites.

Section 5

Page 18792, Line 15: Please add ":" after "in".
Done

Page 18793, Line 21: Perhaps use "masking" or "concealing" instead of suppressing.
Thank you very much for suggesting these words – 'concealing' is used now

Page 18793, Line 23: Please state which scenarios.
Done

Page 18794, Line 4: Please change "found" to "suggested", as this aspect was not fully analyzed, but speculated.
Done.
However, the temperature effect on CH4+OH reaction rate has been discussed in (Patra et al., 2009a).

Page 18794, Line 16: I suggest that the paragraph starting here should be made point (vi) the sequence of points from above, since it describes a conclusion which is fairly central to the paper.
Done

Page 18794, Line 22: Same as above.
Done