Reply to Comments by James Wang

I think this manuscript presents an important intercomparison of model transport characteristics, with a notable component on stratospheric chemistry and circulation and their contribution to surface trace gas concentrations. The paper includes interesting findings and explanations, such as an apparent increase in the rate of interhemispheric exchange over the past two decades and a possible link to a widening of the tropical belt. Also intriguing is the finding that inter-model differences in stratosphere-troposphere exchange may contribute to differences in CH4 growth rate.

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).

There are some areas in which I think the manuscript could be improved:

- The paper would be strengthened with more discussion of the effects of the assumed atmospheric OH abundance on the model distributions of CH3CCl3 and CH4. Something I found quite striking was the overestimate of the interhemispheric (IH) gradient of CH4, and to a lesser extent CH3CCl3, by a majority of the participating models. (The manuscript lacks discussion of possible causes of these discrepancies and the differences among models.) Given that there isn't an overall bias in the simulated IH gradient of SF6, it seems to me that the culprit here might be the assumed emissions, and especially OH, rather than transport. Wang et al. (2008) (full reference below) and Wang et al. (2004) (the reference is in your manuscript) provide insight into this, through extensive analysis of the impacts of OH abundance and interhemispheric distribution and trace gas source strengths on the IH gradients of CH3CCl3 and CH4, respectively. Their inversion analyses resulted in a lowering of global OH abundance and emissions in the Northern Hemisphere extratropics in order to reduce the excessive a priori latitudinal gradient across both hemispheres (they demonstrated that their model IH transport wasn't an issue).

We suspect emissions rather than the OH field to be responsible for the IH gradient discrepancies between measurements and model results (please refer to Figure 9). Models that simulate the SF_6 IH gradient well (within ±0.02 ppt), also do well for IH gradients of CH_4 (within ±4.0 ppb) and CH_3CCl_3 (within ±0.05 ppt). Moreover, models that overestimate the SF_6 IH gradient, also tend to overestimate the IH gradients of CH_3CCl_3 and CH_4. This is clear evidence that the error in the OH-distribution did not play a main role the model-observation mismatches in CH_4 and CH_3CCl_3 IH gradients. Of course OH might play a role in the differences seen for CH_4/CH_3CCl_3 and SF_6, but the main conclusion from figure 7 is that some models are mixing the hemispheres too fast while other models are mixing too slow. Indeed, in GEOS-Chem (used also in Wang et al. 2008) models the IH gradient is modeled quite well.

Another result that leads me to suspect that the specified OH abundance in the current study might be too high is the reported median lifetimes for CH3CCl3 and CH4, 4.61±0.13 yr and 9.99±0.08 yr, respectively. You do note that all of the models except for TM5 have shorter CH3CCl3 lifetimes than those estimated by a number of

previous studies using observed CH3CCl3 (4.9-5.0 yr). You could also cite Wang et al. (2008) here—they estimated even longer lifetimes, 5.0 yr for CH3CCl3 (but note that this also includes a speculative soil sink with lifetime of 45 yr; with respect to tropospheric OH only, the lifetime is 6.9 yr) and 10.1 yr for CH4 (but this includes the soil sink for CH4; without it, the lifetime is 10.9 yr).
The issue of OH abundance could also be discussed in greater depth in the context of the inaccurate simulations of CH3CCl3 annual mean time series in Section 3.2.

By comparing with Prinn et al., which is more of a measurement based benchmark, we suspect the shorter lifetime for CH3CCl3 is arising mainly from its higher loss by photolysis. Note also that the CH4 growth rates are generally well simulated by the model (Fig. 4, top panels) using the same tropospheric OH field. Since we do not run multi-model simulations using varied tropospheric OH strengths or stratospheric loss rates, we cannot comment much on this issue here. However, our results suggest that CH3CCl3 lifetime should be in the range of 4.8-5.0 years, for the assumed emission rates used in this experiment, in order to match the simulated growth rates with the observed one. The models with shorter CH3CCl3 lifetimes tend to simulate smaller growth rates compared to the observed growth rate (ref. Fig. 4 and Table 2).

- It would be helpful if you also provided CH3CCl3 lifetime estimates with respect to tropospheric OH only, and CH4 lifetime estimates with respect to all sinks including the soil sink, for easier comparison with previous estimates including the IPCC assessments.

"…the lifetimes of CH3CCl3 due to photochemical removal is much longer in the stratosphere (~28.6 yr) than in the troposphere (~5.8 yr)…" were given in page#18784, line#10 based on ACTM only because such separation for MCF lifetimes are not available from all models (now TM5 reaults are also included). For CH4, the lifetimes are about 10.2 yr and 186 yr in the troposphere (model levels below tropopause as in ACTM) and stratosphere (model levels above tropopause), respectively. More detailed lifetime analysis has been done using TM5_1x1 model and given below.

Results for the TM5 1x1 run (2000-2005)

(a) CH4_CTL (B = total burden, L = total loss, SL = stratospheric loss, BT = burden troposphere upto 150 hPa, OH = loss OH, OHL = loss tropospheric OH to 150, TS = transport to stratosphere, TL = total loss from troposphere (OH + transport to stratosphere).

CH4-CTL	*B/L	B/OH	BT/OHL	BT/TS	BT/TL
2000	9.67	9.95	8.69	108	8.04
2001	9.67	9.95	8.67	123	8.10
2002	9.64	9.95	8.67	143	8.18
2003	9.63	9.94	8.68	152	8.21
2004	9.64	9.95	8.68	137	8.16
2005	9.62	9.94	8.67	138	8.15

*the difference between this lifetime value and that given in the main paper (10.1 yr) arise from the total burden (TB) calculation. The approximation method used in the paper for B ignores the vertical gradient simulated by different models, a limitation noted already in section 2.5.

(b) CH3CCl3 (L = total loss (ocean + strat + OH), LOH = OH loss, LO = loss ocean deposition, LS = loss stratosphere, TOHL = tropospheric OH loss upto 150 hPa, BT = burden up to 150 hPa, TRS = transport to stratosphere).

MCF	B/L	B/LOH	B/LO	B/LS	BT/TOHL	BT/TRS
2000	4.73	5.78	82.1	38.3	5.17	98
2001	4.72	5.79	82.6	37.2	5.17	109
2002	4.71	5.79	82.6	36.6	5.17	131
2003	4.72	5.78	82.5	37.2	5.17	172
2004	4.72	5.78	82.1	37.7	5.17	137
2005	4.74	5.78	82.4	38.7	5.16	127

Since we did not archive the losses separately for the troposphere and stratosphere, a more detailed analysis will not be conducted. Also some of the differences between lifetime estimates for ACTM (pressure-sigma vertical coordinate) and TM5 (hybrid vertical coordinate) arise from the way troposphere and stratosphere are divided.

• Introduction, lines 14-20: Although you cite a good number of relevant previous CH4 studies, I think you could more precisely and accurately characterize the approaches in the different studies. For example, Wang et al. (2004) not only examined the average “CH4 emission distributions”, but also estimated trends and interannual variations in the emissions and sinks. In addition, Wang et al. (2004) conducted not only “forward modeling”, but also an inversion to estimate the CH4 budget.

We don’t attempt to provide a complete literature overview, but choose to limit the description to what is of direct relevance for our own study.

Reference:
Wang, J. S., M. B. McElroy, J. A. Logan, P. I. Palmer, W. L. Chameides, Y. Wang, and I. A. Megretskaia, A quantitative assessment of uncertainties affecting estimates of global mean OH derived from methyl chloroform observations, *Journal of Geophysical Research, 113*, D12302, doi:10.1029/2007JD008496, 2008.