Supplementary Material

Simulation using CAM-chem model

In order to evaluate our methane emissions on the regional and global scales against atmospheric measurements, we conducted simulations with the Community Atmospheric Model with chemistry (CAM-chem) (Lamarque et al., 2012). CAM-chem is the atmospheric chemistry embedded within the Community Earth System Model (CESM). The model simulations used meteorology input from the NCEP (i.e., the National Center for Environmental Prediction) reanalysis (Qian et al., 2006), allowing particular years and events to be simulated as in a chemical transport model. Details of CAM-chem can be found in Lamarque et al., (2012).

CAM-chem model is one of the 12 models that were involved in a chemistry-transport model intercomparison experiment (TransCom) (Patra et al., 2011). The purpose of TransCom was to investigate the role of surface emissions, transport and chemical loss in simulating global methane concentrations. Seasonal variations of the five CH_4 emission sources were specified: biomass burning (van der Werf et al., 2006), termites (Fung et al., 1991), anthropogenic emissions (Olivier and Berdowski, 2001), wetland and rice paddy emissions(Ringeval et al., 2010;Yan et al., 2009). Monthly mean climatological distribution of OH was used in the TransCom protocol (Spivakovsky et al., 2000) to calculate methane loss.

For this study, we conducted two sets of experiments: one in which we use the TransCom protocol exactly, and one in which we modified the emissions and in particular replaced the wetland and rice paddy emissions from TransCom protocol with those from the CLM4Me′, described in this paper. We compared both sets of simulations against available observations. This provides a preliminary test that the parameterized methane emissions are reasonable. Parameterized wetland emissions have been evaluated within an atmospheric context in Spahni et al. (2011), but not in others (e.g. Wania et al. (2010), Riley et al. (2011), or Ringeval et al. (2010)).

Methods

We repeated experimental setup of the TransCom experiment between the years 1990-1994, but used the wetland and rice emissions derived in this paper. In addition we used a modified version of the fire emissions from the Global Fire Emission Database (GFED version 3 with averaged 21.1 Tg CH_4/yr emitted) (Giglio et al., 2010), instead of the TransCom fire emission taken from GFED version 2 (with averaged 20 Tg CH_4/yr emitted). We also modified the anthropogenic emissions so that the total emissions averaged over the entire period 1990-2007 are identical to those used in the TransCom experiment. As the wetland emissions simulated by the CLM4Me′ model are on the high side of the current estimates (Denman et al., 2007), to obtain a reasonable overall methane source budget the anthropogenic emissions were multiplied by 0.74 while retaining the latitudinal distribution of the TransCom emissions. In the new simulation the total anthropogenic emissions are 217 Tg/year. This is at the low end of estimated anthropogenic emissions, but within the range (209-273 Tg, excluding biomass burning and rice paddies) of reported values in the literature (see IPCC AR4 Chapter 7) (Denman et al., 2007). The distribution of OH used compute the loss of atmospheric methane, as well as termite emissions are identical to TransCom. A comparison of the methane emissions used in this study against those in TransCom is given in Fig.1.

In the following sections, we will specifically focus on comparison of CAM-chem model simulations with our wetlands and rice paddy emissions and with TransCom sources (hereafter, refer to TransCom simulation). Therefore, the difference in model simulations is largely due to different wetlands and rice paddy methane sources.

To initialize the our simulation (not the TransCom simulation), we created the initial condition for atmospheric methane by simulating 1990-1994 period four time using CAM-chem and our modified emission sources as described above. After the fourth time, the methane growth rate in CAM-chem and that in the observations was reasonably close.

Data from ten stations (Table 1) around the world were obtained from the World Data Centre for Greenhouse Gases (WDCGG) at http://ds.data.jma.go.jp/gmd/wdcgg/. We only used monthly data for comparison with CAM-chem and TransCom simulations.

Results

1. Growth Rate

The growth rate was calculated as the mean change of methane concentration from year to year in the simulation period 1990-1994. We also constructed 90% confidence intervals on the mean growth rate (Fig. 2). As suggested in Fig. 2, the mean growth rate of methane concentration in CAM and TransCom is slightly higher than observations except for the high northern latitudes where it is lower. Overall, within the 90% confidence level, the model growth rates derived from this study, from TransCom, and from measurements agree.

2. Seasonal variations in methane concentration

We further compared seasonal variation of methane concentration at these ten stations between model simulations and observations. As shown in Fig. 3, CAM-chem predicted seasonality of methane concentration better than TransCom at the high northern latitude stations of Alert, Barrow, and Cold Bay. At these stations the phasing of the seasonal cycle in CAM-chem is an improvement over that simulated in TransCOM. At all other stations, the seasonal cycle in CAM-chem is similar to that in TransCom.

3. Interhemispheric gradients

Our analysis indicates that CAM-chem overestimates methane concentrations at stations in the Southern Hemisphere (SH) at the beginning and end of these 5 year simulations by ~30-45 ppb (Fig. 4). CAM-chem has consistent higher methane concentration than observations at SH stations during the first year and the last year of the simulation period. This overestimate could easily be corrected by adjusting the initial conditions of the simulation estimated from the model spinup procedure described above.

The interhemispheric gradients (IHG) of methane were calculated as the difference in methane concentration between stations in the Northern Hemisphere (NH) and SH. Fig. 5 suggests that the IHG are higher in TransCom and lower in CAM-chem than observations. However, the seasonal variation of IHG in CAM-chem follows observations much better than TransCom IHG. Particularly, the annual changes in IGH in CAM-chem are similar to observations while TransCom IHG changes are much smaller in magnitude.

Conclusion

We conducted simulation of atmospheric methane concentration in CAM-chem using wetland and methane emissions estimated from CLM4Me′ in addition to other methane sources. We decreased anthropogenic methane sources to the lower end of current estimate in order to have total methane budget matched between this study and TransCom protocol. Observational methane concentration data from ten stations around the world were compared with model simulated methane concentrations as described in the result section. Overall, using our estimated wetland and rice paddy emissions, CAM-chem model performs at least as well or better than TransCom simulation. Thus, within the current level of uncertainty, our emissions appear to be reasonable. Future work will focus on investigating the seasonal variations for the entire period of 1990-2005 and conducting more extensive comparison against observations.

References

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Table 1. Description of the ten stations used in this study

Station Name	ID	Lat	Lon	Elevation (m)	Data availability
South Pole (USA)	spo	89.98S	24.80W	2810	2/1/1983-current
Cape Grim (Australia)	cgo	40.68S	144.68E	94	1/1/1984-current
Tutuila (USA)	smo	14.24S	170.57W	42	4/1/1983-curent
Ascension Island (UK)	asc	7.92S	14.42W	54	5/1/1983-current
Cape Kumukahi (USA)	kum	19.52N	154.82W	3	4/1/1983-current
Mauna Loa (USA)	mlo	19.54N	155.58W	3397	5/1/1983-current
Niwot Ridge (USA)	nwr	40.05N	105.59W	3523	6/1/1983-current
Cold Bay (USA)	cba	55.2N	162.72W	25	5/1/1983-current
Barrow, Alaska (USA)	brw	71.32N	156.60W	11	1/1/1986-current
Alert (Canada)	alt	82.45N	62.52W	210	6/1/1985-current

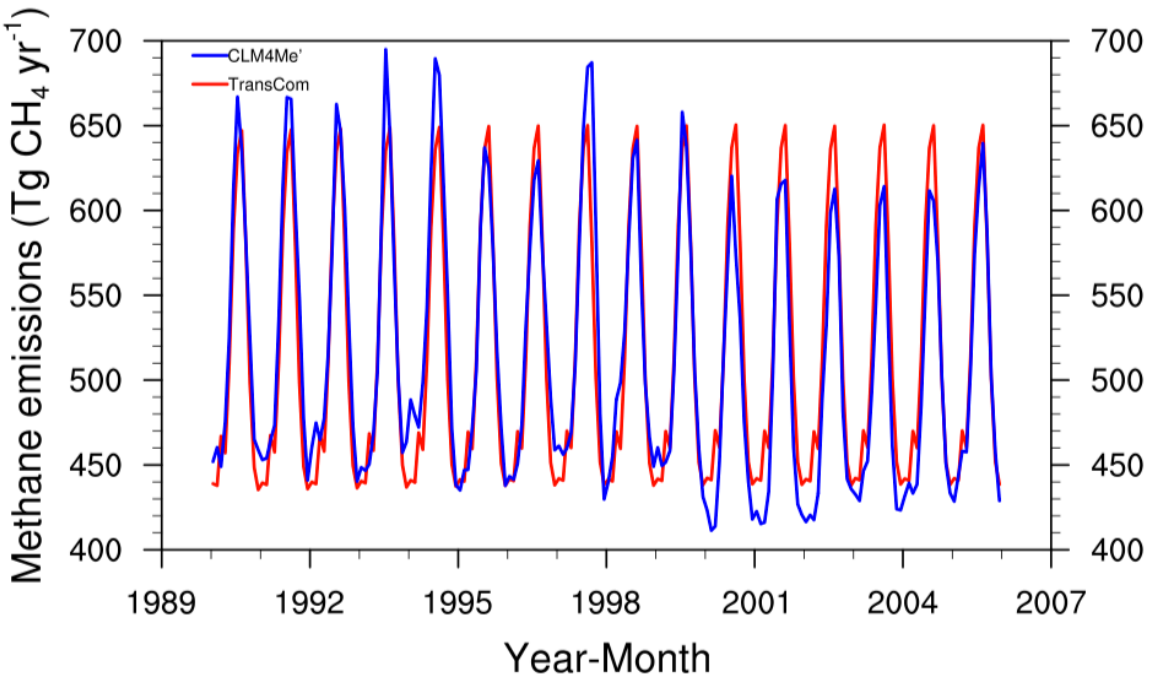

Fig. 1. Comparison of monthly emissions used in CLM4Me′ (this study) and TransCom. The long term annual average methane emissions are the same between CLM4Me′and TransCom. In this diagram, we decreased anthropogenic emissions of methane (still within the range of anthropogenic sources reported in IPCC AR4) in CLM4Me′in order to have total emissions matched between CLM4Me′ and TransCom.

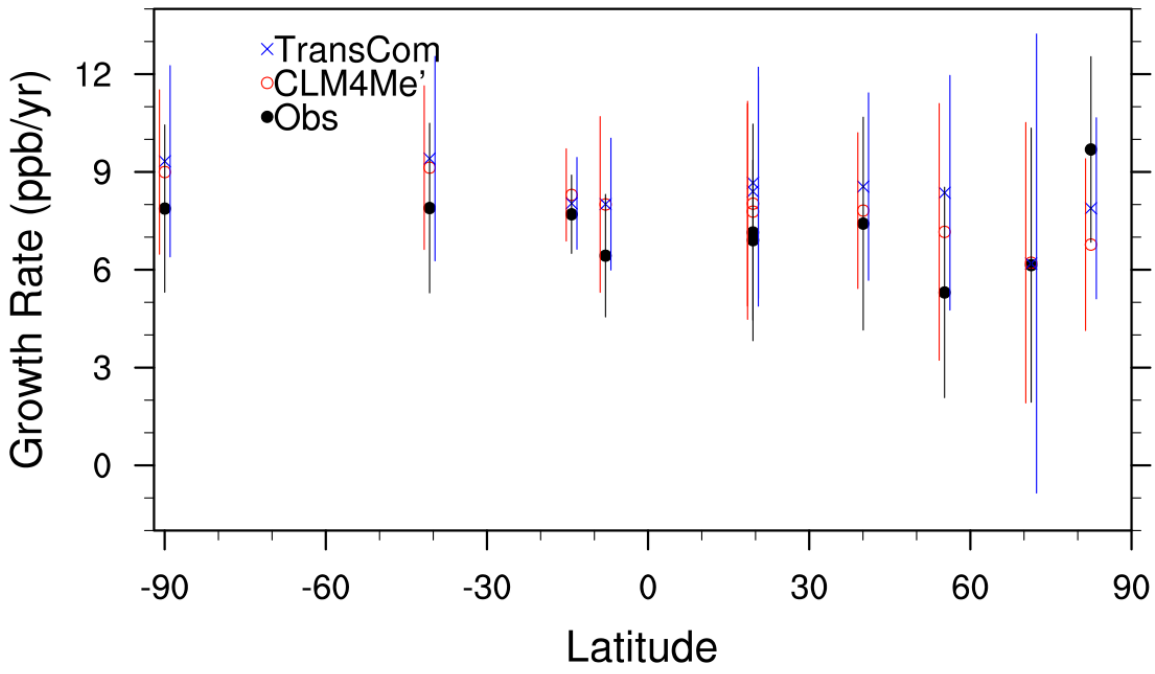

Fig. 2. The comparison of growth rates (ppb/yr) between model simulations and observations. The error bars indicate 90% confidence intervals of the mean growth rate. The confidence intervals of CLM4Me′ (in red) and Transcom (in blue) simulations shifted slightly to the left and to the right in order to distinguish them with confidence intervals of observations at the same stations. The mean growth rates are in dots.

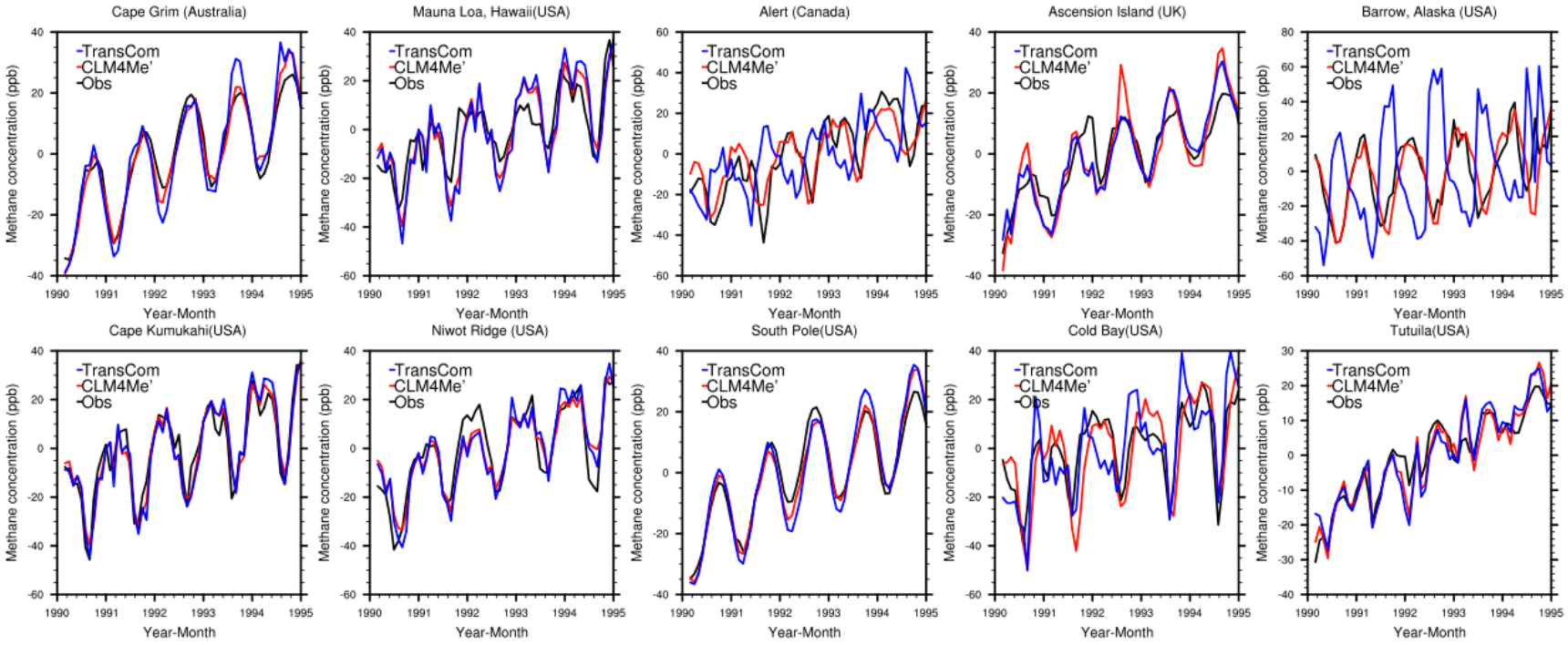

Fig. 3. Comparison of CLM4Me′ (in red) and Transcom (in blue) simulated methane concentration anomalies (with long-term mean removed) with observed anomalies (in black) at these ten ground stations.

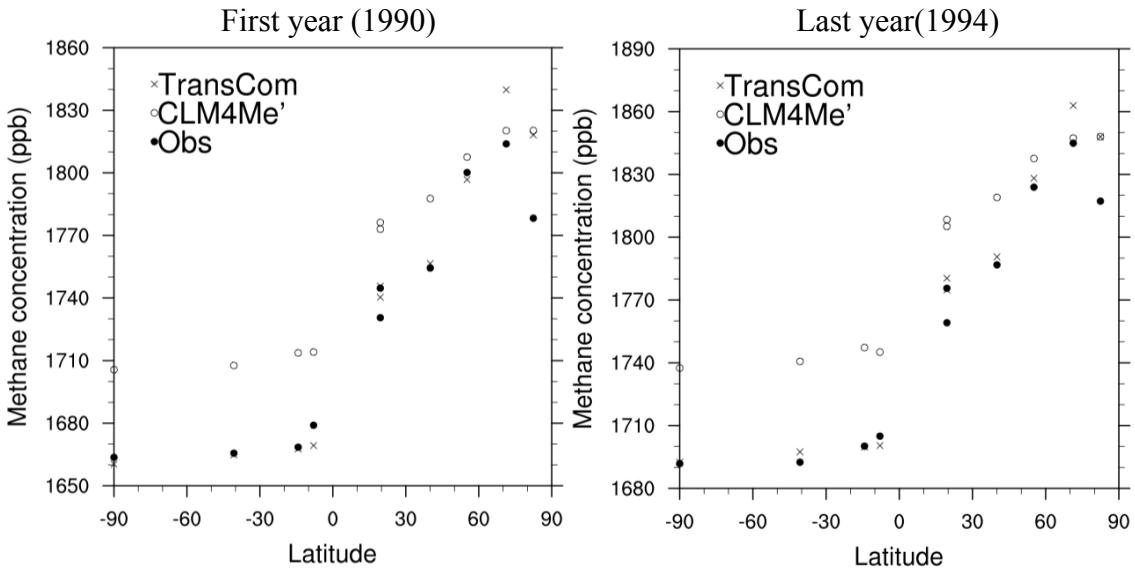

Fig. 4. Comparison of CLM4Me′ and TransCom mean concentrations with observations during the first year and last year of the simulation period.

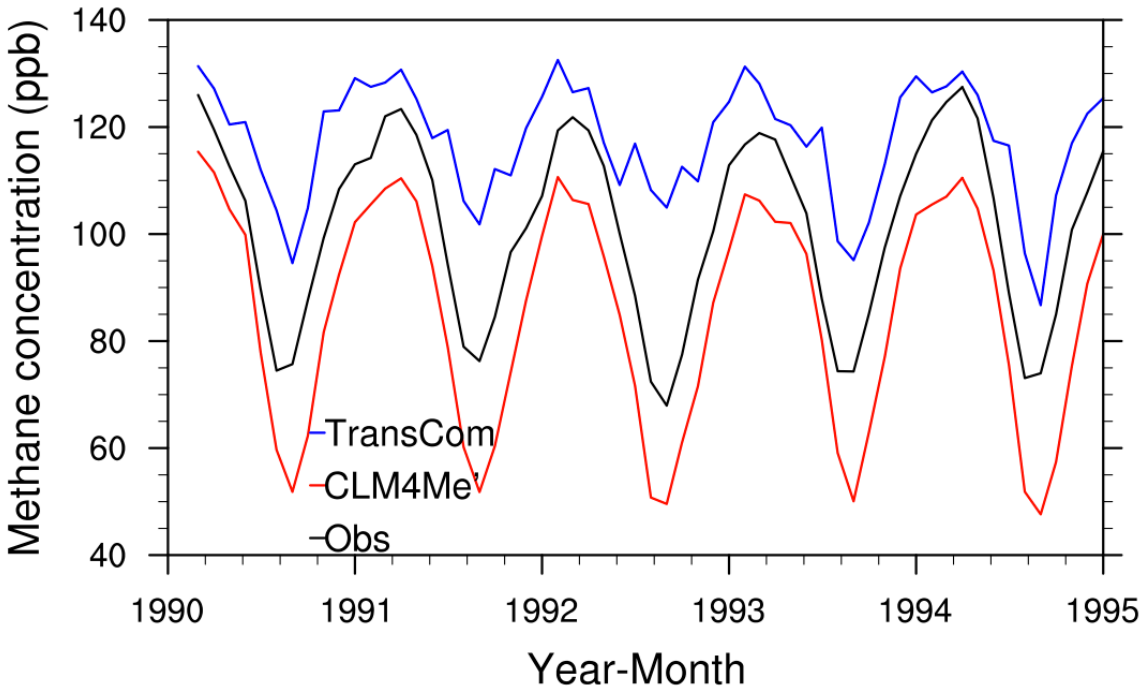

Fig. 5 Comparison of interhemispheric concentration gradients between model simulations and observations. Interhemispheric concentration gradients were calculated as the difference between mean concentrations at the selected stations in the Northern Hemisphere and in the Southern Hemisphere (NH minus SH).