**Atmospheric
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Discussions**

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Interactive comment on "An analysis of atmospheric CH$_4$ concentrations from 1984 to 2008 with a single box atmospheric chemistry model" *by* Z. Tan and Q. Zhuang

Anonymous Referee #1

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The authors simulate global and hemispheric methane concentrations from 1984 to 2008 taking into account various processes (wetland emissions, the other primary emissions, the loss through oxidation by OH and the soil uptake) and the link with CO in a single-box model. Various forward simulations are run to test the sensitivity of CH4 concentrations to the inter-annual variability of OH and to CO concentrations. Two inversions (with a fixed-lag Kalman smoother) contrained by NOAA/ESRL surface measurements of CH4 concentrations are run to optimize CH4 emissions by wetlands.

General comments:

I think the study is below the state of the art. It is nowadays quite possible to study

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Interactive Discussion

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methane concentrations and emissions at the global scale with a spatial resolution of the TRANSCOM regions (see http://transcom.project.asu.edu/T4_methane.php) or even finer, either for forward simulations or for inversions, even for time periods of two decades. Single-box (or two-box) models are used when a large number of processes are taken into account (for example, more reactions in the methane oxidation chain, e.g. with formaldehyde), which is not the case here. Finally, I do not think this study bring anything new to our knowledge of methane trends and its relation with OH and CO variations.

Specific comments:

- observation data: what about the PIs of the measurements used? They are neither co-authors of the paper nor cited in the acknowledgements. Have they been contacted and have they agreed on the use of their data?

- references: a lot of references seem to be out-dated e.g. Etheridge et al. 1992 (see IPCC 2007), Thompson et al. 1986. Some recent studies seem not to have been used e.g. Bousquet et al. 2011 regarding methane source attribution.

- section 3.1: the text is not always clear on the hypothesis that is currently commented (e.g. constant or variable OH concentrations).

* Section 1:

- p. 30261, l. 5: "surprisingly". This has been published, so why be surprised?

- p. 30261, l. 15: "as the largest natural source of methane, emissions from wetlands are very sensitive to environmental factors". Other sources are also sensitive to environmental factors, even anthropogenic sources (e.g. rice paddies).

* Section 2:

- p. 30262, l. 9: "earlier studies". Which ones?

- p. 30263, l. 12-14: "as isoprene oxidation has been suggested to serve as both a

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sink and source for hydroxyl radicals (Taraborrelli et al., 2012), it is necessary to also include OH production from this pathway into OH sources" Isoprene is one of the VOCs that is oxidized by OH. In the rest of the text, isoprene is not mentionned anymore, only "VOCs". Could the partition between isoprene and other VOCs be made clearer?

- p. 30263, l. 19: "only wetland methane fluxes show clear evidence of sensitivity to climate change". What of other processes taken into account here, such as the emissions of CO (and CH4) due to biomass burning or the emissions of CO due to fossil fuel consumption? Could you quantify the sensitivity to climate change of the various processes to justify holding SCH4 and SCO constant?

- p. 30263, l. 26: "by adding a constant southward transportation item to Eq. (1)". I think there is no other information on the choice and magnitude of this item, could you give more details?

- p. 30277, Table 1: Why not indicate the distributions of wetlands, methane oxidation, CO emissions and oxidation, production of OH by ozone photolysis (the Table is "Distribution of sources and sinks")? All the figures given in the caption lack references (and some necessary to the model are missing, e.g. CO emissions,...). Given the difference in land masses, why are the distributions for CO and OH 50/50? Shouldn't the difference in land areas imply for example a difference in the distribution of VOCs and therefore, in OH? Regarding CO surface deposition, what is the reference for the 290 Tg/y? Shouldn't also the deposition be different in the two hemispheres due to the difference in land surfaces?

- p. 30264, l. 6-7: "the initial concentrations of CH4, CO and OH". Where do these initial concentrations come from? A spin-up of the model? Observation data?

- p. 30264, l. 15-16: "we calculate methane fluxes from the Northern and Southern Hemisphere separately and then sum them up together." compared to p. 30270, l. 6: "We also ran our model under the constant OH assumption in the Northern and Southern Hemispheres, respectively." It is not clear for me whether, for each forward

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Comment

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Interactive Discussion

Discussion Paper

or inverse result, there are two runs, one for NH and one for SH and the global results are the sum of there or there are three runs, one for the global and one for each hemisphere. Could you state this more clearly?

- p. 30266, l. 6-8: "state variables" and "fluxes". Could you state more clearly what is in your state vector? I think there are monthly CH4 fluxes due to wetlands but maybe there also are the initial concentrations?

- p. 30267, l. 14-15: "Then, for the correlations between this flux and fluxes from J-1 to J-5, they are calculated by analyzing the time series of the Q10 function in Eq. (1)." How are the time series analyzed? What correlations are retrieved?

- Section 2.3: I understand that only CH4 concentrations are constrained in the inversion. Why not constrain also CO concentrations and even, OH concentrations (with a proxy such as methyl-chloroform as is often done)?

* Section 3:

- p. 30268, l. 7-10: "Because the concentrations of hydroxyl radical decline from 10 to 9.76×10^5 molecule.cm^{-3} in the experiment, this slowdown is more likely a result of the stabilization of methane emissions, which was caused either by weakened global warming or by constrained anthropogenic emissions." The variations in OH concentrations are not necessary solely due to its reaction with methane: there may be numerous other causes. There may be also a number of other causes to the stabilization of methane emissions, such as a balance between two different causes, and they do not necessary have a (direct) link to global warming. Why focus on these causes?

- p. 30268, l. 10-12: "Our simulation also implies that climate change is responsible for at least one methane concentration anomaly in recent decades: the abrupt global methane increase during the El Nino events of 1997 and 1998 (Dlugokencky et al., 2001)." Why does the simulation imply a direct link between climate change and the El Nino event of 97-98?

- p. 30268, l. 17: "this bias". Is it acually a systematic bias computed over 2000-2008?

- p. 30268, l. 17-19: "We think one possible reason for this bias is that the methane emissions from non-wetland sources, such as the natural gas industry, decreased beginning in 2000. However, we lack emission data to validate this hypothesis." Is there really no emission data to check this hypothesis? At least over some industrialized regions, it would probably be possible to use anthropogenic inventories (e.g. EDGAR4 over Europe) or even optimized emissions from independent studies.

- p. 30268, l. 24-26: "Furthermore, if atmospheric OH declines with time, CO abundance should increase accordingly for the attenuation of oxidation power, as in our model, where the simulated CO rose from 93 to 96.9 ppb." This is only valid if CO emissions do not also decrease with time.

- p. 30269, l. 14-17: "In the model, simulated [CO] increases from 120 to 125.2 ppb in the north and from 66 to 68.6 ppb in the south, and meanwhile simulated [OH] decreases from 10 to 9.73×10^5 molecule cm^{-3} in the north and from 10 to 9.8×10^5 molecule cm^{-3} in the south." How does this compare to the observations (at least for CO)? What can be deduced from that?

- p. 30270, l. 5: "In this simulation, atmospheric CO increases from 93 to 95.6 ppb." How is this linked to methane variations?

- p. 30270, l. 13-14: "wetland and rice agriculture emissions (both sensitive to climate change)" See specific comment to p. 30263, l. 19: this is not compatible.

- p. 30270, l. 19-21: "In the simulations, modeled [CO] increases from 120 to 122.9 ppb in the north and from 66 to 68 ppb in the south. " Same as specific comment to p. 30270, l. 5.

- p. 30270, l. 26: "considerably". Quantify.

- p. 30270, l. 28: "the buffer effect of VOC oxidation". What is meant here? VOC oxidation covers a large range of life-times.

- p. 30271, l. 2-4: "So we guess that a mechanism to stabilize OH concentrations should exist, and the lack of formulation of this mechanism in our model has caused the discrepancy in Fig. 4a." OH is lost in many reactions so that there are many other explanations possible for the discrepancy.

- p. 30271, l. 17-20: "Because other methane sources besides wetland emissions are set as constant, changes in methane fluxes in other ways would be also attributed to wetland incorrectly, for example, the peak in 1991 should be attributed to the eruption of Mt. Pinatubo." This is a strong limitation to the results of the inversion where only wetalnd emissions are optimized. Why not optimize other terms of Eq. (1)?

* Section 4:

- p. 30271, l. 22-25: "Using a simple one-box atmospheric chemistry model involving CH4, CO and OH, we can more efficiently re-construct atmospheric methane con-centration trajectory in recent decades in comparison with using 3-D transport and chemistry models (Patra et al., 2011)" This is far-fetched. In modelling, one can always get the "right" results for the wrong reasons. The real point is the knowledge of the modelled mechanisms, on which the TRANSCOM-CH4 community has been working for several years. I therefore think they would have noticed if a simple one-box model would do the trick to explain such a complex issue.

- p. 30272, l. 1-2: "there should be other sources or sinks that can influence methane concentrations significantly, but if OH is constant during our simulation period, atmo-spheric CH4 can be predominantly determined by these two factors. " This is only true in this model.

- p. 30272, l. 11-12: "we estimate the global wetland methane emissions to be ap-proximately 128 Tg/yr." Why give a mean when the aim of the inversion is to retrieve fluxes with a monthly (or yearly?) time-resolution, including the trend and inter-annual variability? A figure displaying the retrieved variability of wetlands would be more infor-mative.

Technical corrections:

- p. 30263, l. 25: "to order to" -> in order to?

- p. 30264, l. 2: "earth" -> Earth?

- p. 30267, l. 14-15: "Then, for the correlations between this flux and fluxes from J-1 to J-5, they are calculated by analyzing the time series of the Q10 function in Eq. (1)." This sentence seems a bit strange to me.

- p. 30267, l.18: "estimate" -> estimated?

- p. 30268, l. 16: "reaching 20 ppb higher than" -> being 20 ppb higher?

- p. 30268, l. 25: "for the attenuation of oxidation power" -> due to the attenuation?

- p. 30269, l. 8: "at the same level of observations" ->at the same level as?

- p. 30269, l. 25: "shows that methane growth rate" -> shows that the methane growth rate?

- p. 30270, l. 2: "are not well presented" -> are not well represented?

- check when capitals are needed at "north" and "south" through the text.

- p. 30270, l. 15-16: "In the South, as its most areas are covered by oceans, " -> as most of its surface is covered by oceans?

- p. 30270, l. 18: "transportation" -> I think transport is meant here.

- Letters used to label the sub-figures (a, b, ...) do not appear in the Figures.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 30259, 2012.