Interactive comment on “Seasonal Variability of Stratospheric Methane: Implications for Constraining Tropospheric Methane Budgets Using Total Column Observations” by K. M. Saad et al.

Anonymous Referee #2

Received and published: 7 June 2016

General comments

This paper by Saad et al., compares the agreement between Total Carbon Column Observing Network (TCCON) and a chemistry transport model (GEOS-Chem) for total and tropospheric column-averaged mole fractions of methane, the second anthropogenic greenhouse gas. From this comparison they infer possible consequences on the methane emissions as estimated by atmospheric inversions using chemistry transport models. The main results include the largest discrepancies between model and observations in the Northern hemisphere stratosphere increasing with latitude as the
tropopause height decreases, and a lag in the model's tropospheric seasonality most probably driven by transport errors. One interesting results is that these errors partly compensate in the total column of methane indicating the possibility to get reasonable agreement for total columns with a wrong vertical transport. This work has implications for atmospheric inversions although the precise quantification of the impacts of the errors found in this paper remain partly to be done. It addresses an important matter as transport model errors are the 2-nd largest cause of uncertainty, after observations space and time density, in atmospheric inversions. Many papers have addressed impact of pbl (rectifier effect) or large-scale horizontal transport (e.g. inter-hemispheric exchange time) but less the impact of vertical transport (e.g. Locatelli et al. 2015).

Nevertheless, the paper needs attention before publication in ACP. It lacks precision in the text in many places (see specific comments), so are legends of some figures. Several important sentences, often when synthetizing results are confusing and not clear and make the reading not fluid at all with this version (see specific comments). I find the result section, a bit too descriptive, not providing systematically explanations or hypotheses for the inferred results. This has to be improved as it is not done either in the discussion part. About the hypotheses, for instance among several other things reported below, I wonder why the aseasonal run disable the seasonal emissions and scale up the rest instead of prescribing the annual mean of seasonally changing sources ? This is strange as it changes the spatial distribution of emissions on the top of the suppression of seasonality. Also, the implication for atmospheric inversions should be more clearly expressed in the discussion section.

Specific comments

Abstract : “ large number of highly variable sources Âž not all methane source are highly variable. On what scale ? And sinks ? I suggest because of a large number of uncertain sources and sinks.

Page 2 : lines 1-5 : the words Â’n atmospheric inversion Âž should appear somewhere
in this paragraph. Lines 16-17: Do they have the same bias as aircraft observations of clear-sky only measurements (aircraft do not fly in bad weather conditions)? It is worth noticing this issue somewhere. Lines 18-20: Fraser et al: how did they do that? did they account for observation systematic errors as well? Please be more precise when quoting papers. Idem for Wecht et al. Lines 33-35: ambiguous sentence. Please rephrase. Indeed tropospheric CTM do not reproduce well stratospheric transport.

Page 3: Line 6: systematic model biases: strange expression. Maybe systematic errors would be enough. What about the random part or errors? Do you address this as well? Please reformulate. Line 6: "seasonal cycle and spatial distribution of CH4 emissions? please be more precise. Line 15: it would be good to briefly recall how the TCCON total columns are inferred. In particular, what is the influence of the modelled CH4 profile used in the retrieval (as a prior) on the final product. As this profile comes from a model, it would be worth commenting on this considering the topic of the paper. L16: precise: please be more quantitative here or remove the word. How precise compared to surface networks for instance? how is your data uncertainty estimated?

Page 4: Lines 14-15: please provide a reference for emissions and OH. Do they vary inter-annually? For OH concentrations, what is your ratio NH/SH? More precisions are needed here. Indeed you release emissions every hour but their time evolution is monthly or annually probably. Please precise this not to let the reader think that we know methane emissions with an hourly time step!

Section 2.2: It would be useful to position GEOS-CHEM with other transport models based on previous Transcom-like experiment (e.g.: Patra et al., 2011): is it a "fast" model (inter-hemispheric exchange time?), what about stratosphere/troposphere exchange time? ... It would be very useful for other modellers to use the results of the paper.

Page 5: Line 1-2: this first sentence needs precision: what is GGG2014? What
is GEOS5? Acronyms have to be defined and explained Lines 10-15: the choice to disable the seasonal emissions and scale up the rest is strange as it changes the spatial distribution of emissions on the top of the suppression of seasonality. Why not prescribing the annual mean of seasonally changing sources? Line 15. What is ‘TCCON daily median scaled priors Â¿?’ you need to provide more details here. What is the influence of these Â¿ priors Â¿ on the TCCON products and on the comparison proposed here. Line 20: Â¿ While XtCH4 20 changed slightly Â¿: how much is the change? please provide % for instance. Why only testing above levels? please provide explanations. Line 24: "small". Please be more precise. Remain within ±5 ppb for instance? Idem for larger NH changes: ‘varies from -10 to +13 ppb’.

Page 6: Lines 1-2: what do you mean by Â¿ common Â¿? Why the age of air increases when seasonality is suppressed? Please provide more clear explanations. Line 3: “relatively short Â¿: please provide an estimate

Page 7: Line 9-10: tropospheric slope does not seem lower than one for southern stations. Indeed it seems there is a little north-south gradient in the tropospheric slopes. Did you investigate it?

Figs 4: this figure is not enough analysed. You do not comment: - the negative bias of GEOS-CHEM at most sites for the tropospheric & total columns (4ab) - the fact that stratospheric columns of GEOS-CHEM seems underestimated for more southern sites and overestimated for more northern sites (4c) - possible reasons for the poorer agreement in the stratosphere. You may also consider two slopes, one for the southern stations (larger than 1) and one for the northern stations (smaller than 1) on fig 4a, or a non linear continuous decrease of the slope from south to north. Why only keeping a global slope?

Page 8 Line 3-6: any possible explanation for the differences with ACE? Line 12-14: “As the effective..pressure heights”: unclear sentence. Please rephrase. Page 10 line 10: Â¿ production Â¿ or emissions?
Page 8-9 Line 15-4: the part about troposphere is confusing as figure 6b shows similar trend for stratosphere and troposphere but you mention in the text much lower sensitivity. Please clarify this section.

Page 10 Line 5: “production” do you mean emissions as there is no methane 3D production in the atmosphere? More, your statement brings more the summer large wetland emissions as an explanation for the phase of the modelled signal than the loss which should produce more a fall maximum as in surface observations (although Par falls is not the best example to discuss seasonal variations as the signal is complex). Please clarify.

Page 11: Lines 1-3: please develop a bit why you discard OH as an hypothesis to explain the inferred changes? Lines 7-8: “The model sensitivity kernel implicitly includes .. Âž well do you mean variance matrices associated with observations? with prior emissions? Indeed, transport errors are generally implicitly include in atmospheric inversions by inflating observations errors but are not part formally of the variance matrix of emissions. Lines 7-8: “which are compounded if vertical levels are subject to different errorsÂž Confusing sentence. What do you mean? Please clarify.

Page 12: Line 6 ‘Although the stratosphere accounts for about 30% ‘ if you refer to top panel of figure 9, I suggest up to 35% (JJA) Fig9: The legend of figure 9 is unclear. Top panel: fraction of what? Bottom panel: the orange curve is a difference or the error of the aseasonal? Unclear. line 10: “The seasonality of the stratospheric error will therefore distort the inversion mechanism and thus posterior emissions estimates. Âž: well only if these error are not included in the inversion variance matrices. I would be more confortable writing may distort or precise the conditions of influence of the seasonality in the stratospheric signals on surface emissions through inversions. Line 11: Â„ product of transport errors Âž: how did you evaluate the possibility of issues related to OH radicals? Lines 10-12: it is never mention except in caption of figure 4 that ‘t’ in CH4t refers to troposphere. “their emissions are very uncertain Âž: you may quote a recent estimate such as in Kirschke et al. 2013 or IPCC.
Page 13: Lines 7-8: “both the magnitude and seasonality of the difference is significant. $\dot{\alpha}$: the unit (tons) makes it difficult to say so. There is obviously a sensitivity if transport error shift the seasonality but what does it give in terms of ppb? or in terms of % of initial emissions? This would be more clear for the reader. Lines 8-10: “The largest disagreements between measured and modeled $X_t$ occur ... than annually. $\dot{\alpha}$ This sentence is unclear to me. Please rephrase.

Page 14: line 16: “the meridional gradient $\dot{\alpha}$ of what? emissions? concentrations? Unit of figure 12? Kg/yr? Maybe change to Tg/gridbox or Tg/yr/°latitude?

Conclusions Line 3: re-precise in the start of conclusion the you used GEOS-CHEM and what are XCH4 and XtCH4 as it has to be readable by itself.

Page 15, lines 1-5: If stratospheric ch4 is largely independent from tropospheric CH4, is it worth developing full tropospheric and stratospheric chemistry models or prescribing stratospheric CH4 based on satellite observations is enough?

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-303, 2016.