

**We would like to thank Anonymous Referee #1 for the valuable comments. Below are our responses in blue:**

Modelling of chemical composition of the troposphere has been challenging in particular for the short-lived species, such as the hydroxyl (OH) and its feedback with the green- house gases (e.g., methane) and other air pollutants (e.g., CO). The authors show a simplified approach to get reasonable answers to these questions, which are welcoming news for the community. The CH<sub>4</sub>-CO-OH system has now been implemented in the NASA GEOS-5 Atmospheric Global Circulation Model for chemistry-climate feed- back related studies. However, the challenges remain on the validity of this simplified CH<sub>4</sub>-CO-OH approach as that is the case for the state-of-the-art full chemistry model. The manuscript is generally well prepared, and can be published after minor revisions. My specific comments are listed below.

**p.9453, line 24-26: Agree, but the system is so non-linear that this simplification may bias the results in different ways! So I am not sure whether you can claim that CH<sub>4</sub>-CO- OH will be any better.**

We do not claim that ECCOH is “simplified”, but rather “computationally efficient” - the main advantage of our code. To address this concern, we have considerably “beefed up” our description of the ECCOH module in Section 2.1.

p.9454, line 19-21: Can you be a bit more specific here - are you talking about the global mean OH or also about the inter-hemispheric distribution or regional differences? If all, it would be useful for the readers to get some of your opinions on each of the issues. Also relevant for the paper for discussions later on.

We modified the sentence from:

“Furthermore, simulated OH from full chemistry mechanisms in global models is still highly uncertain because of incomplete knowledge and representation of OH sources, sinks and recycling (e.g., Elshorbany et al., 2010, 2012a, b, 2014; Stone et al., 2012).”

To:

“Furthermore, simulated OH from full chemistry mechanisms in global models is still highly uncertain because of incomplete knowledge and representation of OH sources, sinks and recycling (e.g., Elshorbany et al., 2010, 2012a, b, 2014; Stone et al., 2012). For example, 1) nitrous acid (HONO) is typically underestimated in models by an order of magnitude (Elshorbany et al., 2012b), which can lead to a significant underestimation of OH, especially in urban high-NO<sub>x</sub> regions; 2) in unpolluted, forested environments, significant discrepancies exist between models and measurements (Stone et al., 2012); and 3) Patra et al. (2014) indicate that the inter-hemispheric OH ratio (northern to southern hemisphere) is near unity, while a recent model inter-comparison had a multi-model average of about 1.3.”

Patra, P. K., Krol, M. C., Montzka, S. A., Arnold, T., Atlas, E. L., Lintner, B. R., . . . Young, D. (2014). Observational evidence for interhemispheric hydroxyl-radical parity. *Nature*, 513(7517), 219-23.

p9458, line 6-7 : add 'et al.' to Lawrence. Also I think MCF is used as a proxy for OH since the 1990s by many others. Generally agree, but MCF has recently been used at quite good confidence for broad characterization of OH in the two hemispheres because uncertainties MCF emissions are now small compared its atmospheric burden.

We added the following text (underlined) to the paragraph in question:

“There are very few direct observations of OH with which to constrain models (e.g., Stone et al., 2012) and none on regional or global scales. Therefore, the methylchloroform (MCF) lifetime inferred from measurements serves as a widely used, indirect proxy for global OH abundance (e.g., Lawrence, 2001). Though useful, the MCF lifetime gives an incomplete description of the spatial and vertical distributions of OH (e.g., Lawrence et al., 2001) and there are uncertainties concerning MCF emissions and the resulting lifetime estimate (e.g., Wang et al., 2008). Nevertheless, the MCF data have been recently used to infer the ratio of OH in the Northern to the Southern Hemisphere (Patra et al., 2014).”

p9458, line 18 : can you be a bit more quantitative here? what is quite?

We added the following text (underlined) to the sentence in question:

“The seasonal and spatial distributions of the zonal mean OH in the *Base* scenario are quite comparable to the OH climatology of Spivakovsky et al. (2000), despite the different inputs given to the parameterization of OH in the two studies.”

p9460, line 5 : I think very few sites were in place in 1980s, thus this sentence is misleading. Can you not arrive at this conclusion by using only the sites with full data coverage?

To address this concern, we modified Figure 4 and modified (underlined text) the first paragraph of Section 3.2 to:

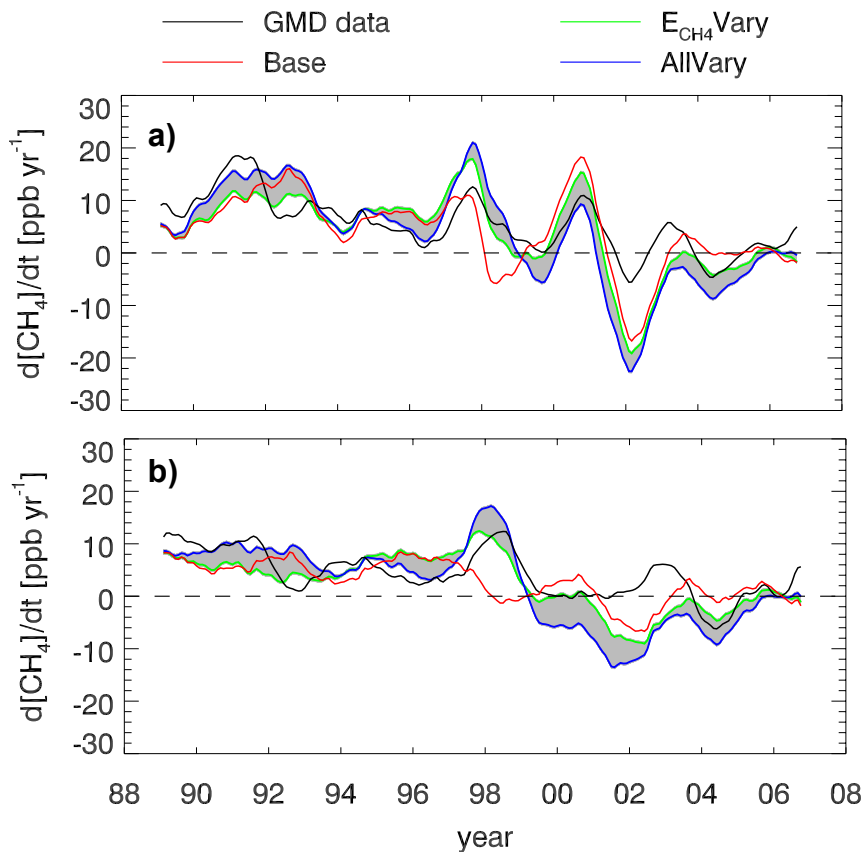


Figure 4: a) 12-month running mean atmospheric growth rate of methane ( $\text{ppbv yr}^{-1}$ ) for the average of 92 GMD stations and from model output for several scenarios averaged for those station locations. The shaded area is the difference between the  $E_{\text{CH}_4}\text{Vary}$  and  $AllVary$  scenarios, which indicates the total contribution of nonlinear feedbacks (i.e., from variations of CO emissions and variables input to the parameterization of OH) of the  $\text{CH}_4\text{-CO-OH}$  system to methane's growth rate. b) Same as a) but for the average of 17 GMD stations, which covers 100% of the simulation period.

*“GMD surface data:* We evaluate our simulated surface distributions of methane from the *Base* scenario with data from the NOAA Global Monitoring Division (GMD) network. The simulated, interannual variation of methane's global growth rate agrees reasonably well ( $R^2 = 0.44$ ) with that estimated from GMD data, using all available data from 92 stations over the simulation period 1988-2007 (Fig. 4). We decided to include all 92 stations, even those without records that cover the entire simulation period, as we are able to nearly reproduce Fig. 4 using 46 stations that have at least 75% data coverage (Fig. S 4). The agreement of model output with observations is worse ( $R^2 = 0.33$ ) when we only use the 17 stations that have records covering the entire simulation period (Fig. 4). A relatively high correlation coefficient ( $R^2 = 0.44$ ) implies that interannual variations in anthropogenic methane emissions and dynamics explain much of methane's growth rate over the study period, which is consistent with the findings of the TransCom model intercomparison project (Patra et al., 2011).”

p9460, line 14 : Please mention the time period of the data used

Done. The simulation period is 1988-2007.

p9460, line 24 : I thought the base case doing well as seen in Fig. 5, and this problem is more serious in All Vary case

The sentence in question refers to only the *Base* scenario, so the statement is true.

In section 4.3, we compare the *Base* and *AllVary* scenarios to observations, discussing in what regions/time periods the scenarios perform best/worst.

p9462, line 12 : Are those in the tropics or SH?

We added the following text (underlined) to the sentence in question:

“However, the largest biases (Figs. 12 and S14) occur over (1) tropical and subtropical biomass burning regions (~20 %) during boreal winter, indicating that either the CO emissions used in the Base scenario are too high or that simulated OH is too low, and (2) most of the Northern Hemisphere (< -20 %) during the summer season, indicating that either CO emissions are too low or that OH levels are too high, which is consistent with previous studies using similar emissions (e.g., Shindell et al., 2006; Strode et al., 2015).”

p9462, line 21 : But in your model, you see these biases also at the surface stations, in the previous para - right? So the uncertainties in remote sensing products may not be questioned in this context.

The simulated near-surface concentration of CO in the *Base* scenario is underestimated, especially in the northern hemisphere related to using annually repeating CO emissions. Using annually-varying CO emissions (e.g., *FFBBE<sub>CO</sub>Vary* and *AllVary* scenario) solves this issue.

For MOPITT and TES/MLS, the comparisons reveals higher simulated CO over biomass burning regions during boreal winter and lower simulated CO over the northern hemisphere during summer. The comparison results are consistent with previous model-MOPITT comparisons as mentioned in the text.

p9464, line 10 : The problem is how to say something meaningful out of this comparisons. Can we claim Voulgarakis et al. (2015) is right?

Our results support the conclusions of Voulgarakis et al. (2015) and it also makes sense given that CO interannual variability is a strong function of variations in CO emissions.

p9465, line 12 : I have some concerns when the MCF-inferred OH IAV is marked as "Observation" in Fig. 15. A lot of assumptions has gone in this calculation, so I would recommend to change the legend as 'MCF-inferred" or something like that.

Done.

p9466, line 3 : The fires also emit a lot of ozone precursors. If O<sub>3</sub> increases will there be more OH production under no/less cloudy conditions - how does those feedback works? Any perspective will be appreciated.

It is true that other OH/O<sub>3</sub> precursors are emitted from biomass burning. For example, we know that large amounts of HONO, a primary OH source, are emitted from fires (e.g., Yokelson et al., 2009). However, these fire plumes are characterized by low NO<sub>x</sub> conditions and thus OH will not be efficiently recycled (as per current knowledge), which may limit the impact of these precursors on OH and O<sub>3</sub>. It has also been shown in other studies that that high boreal fire activity increases O<sub>3</sub> in the free troposphere by up to 10 ppbv with photochemically aged biomass burning influencing tropospheric oxidant chemistry after 1-2 weeks of transport to the region (Parrington et al., 2013 and references therein). However, the extent and the mechanism of these impacts are still not well known. Therefore, it is not possible to test these possible impacts in global models without knowing the actual mechanisms.

Yokelson, R. J., Crouse, J. D., DeCarlo, P. F., Karl, T., Urbanski, S., Atlas, E., Campos, T., Shinozuka, Y., Kapustin, V., Clarke, A. D., Weinheimer, A., Knapp, D. J., Montzka, D. D., Holloway, J., Weibring, P., Flocke, F., Zheng, W., Toohey, D., Wennberg, P. O., Wiedinmyer, C., Mauldin, L., Fried, A., Richter, D., Walega, J., Jimenez, J. L., Adachi, K., Buseck, P. R., Hall, S. R., and Shetter, R.: Emissions from biomass burning in the Yucatan, *Atmos. Chem. Phys.*, 9, 5785-5812, doi:10.5194/acp-9-5785-2009, 2009.

Parrington, M., Palmer, P. I., Lewis, A. C., Lee, J. D., Rickard, A. R., Di Carlo, P., Taylor, J. W., Hopkins, J. R., Punjabi, S., Oram, D. E., Forster, G., Aruffo, E., Moller, S. J., Bauguitte, S. J.-B., Allan, J. D., Coe, H., and Leigh, R. J.: Ozone photochemistry in boreal biomass burning plumes, *Atmos. Chem. Phys.*, 13, 7321-7341, doi:10.5194/acp-13-7321-2013, 2013.