Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-529-AC1, 2019

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Investigation of the global methane budget over 1980–2017 using GFDL-AM4.1" *by* Jian He et al.

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Received and published: 18 October 2019

We thank the reviewer for the insightful comments and suggestions on our manuscript. Below, our responses in bold text follow the reviewer's comments shown in plain text.

General comments: The work describes CH4 budget using an atmospheric chemistry-transport model that is developed at the GFDL. The authors have taken in to account all possible causes of variabilities in CH4 budget, such as the emissions and loss due to tropospheric hydroxyl (OH). As shown in the manuscript, OH variability is of as much importance as the emissions in explaining the CH4 growth rate variabilities in different decades in the period of 1980s to 2010s. The manuscript is generally well

written. However, I felt toward the end of the manuscript is a bit of stretch and could be reduced (I have made some suggestions in my specific comments). The manuscript can be accepted after a major revision.

Reply: We thank the reviewer for the positive comments. As suggested by the reviewer, we have shortened Section 3 and revised the manuscript carefully. Below are our point-by-point responses.

Specific comments: Line 49 62ff: can the growth rate discussions in the introduction be made concise and put together at one place.

Reply: As suggested by the reviewer, we have removed the sentence highlighting past studies on drivers of methane trend and variability avoid redundancy. However, we keep the sentence on the observed changes in growth rate to motivate the study.

Line 80ff: I think there are other prominent inverse modelling results trying to explain the recent regrowth of CH4 concentrations.

Reply: We have revised this sentence and included additional references in the revised manuscript as below:

"The observed renewed growth since 2007 has been explained alternatively through increases in tropical emissions (Houweling et al., 2014; Nisbet et al., 2016) such as agricultural emissions (Schaefer et al., 2016; Patra et al., 2016) and tropical wetland emissions (Bousquet et al., 2011; Maasakkers et al., 2019), increases in fossil fuel emissions (Rice et al., 2016; Worden et al., 2017), decreases in sources compensated by decreases in sinks due to OH levels (Turner et al., 2017; Rigby et al, 2017), or a combination of changes in different sources such as increases in fossil, agriculture, and waste emissions with decreases in biomass burning emissions (Saunois et al., 2017)."

Line 135-137: This is a quite strange statement. After reading the whole manuscript I

do not believe you have tried to address these couple of issues to a great extent. May be remove?

Reply: We have removed these sentences as suggested by the reviewer.

Line 156: Not from wetchart? I mean does wetchart not have IAV?

Reply: We use WETCHARTs version 1.0 (Bloom et al., 2017) for wetland emissions. The seasonality and spatial distributions are based on ensemble mean. We then repeated the emissions every year. There is also an extend ensemble version of WETCHARTs with interannual variability of wetland emissions only for 2001-2015. We did a sensitivity test with this version to compare with climatological wetland emissions. The results are shown in Figures S2 and S3 in the Supplement. We find a better model performance with climatological wetland emissions. Besides, with the optimization on wetland emissions, the signal of the initial interannual variability of wetland emissions would be lost anyway. Therefore, we keep this version (i.e., climatological wetland emissions) as the starting point for wetland emission optimization.

Line 206: Not clear if this is after LNOx scaling? please make this statement precise (e.g., Control).

Reply: This magnitude is for standard AM4.1 without scaling. We have revised the sentences in the revised manuscript to make it clearer as below.

"The climatological global mean LNOx emission simulated by standard AM4.1 is about 3.6 TgN yr-1, within the range of 2-8 TgN yr-1 estimated by previous studies (e.g., Schumann and Huntrieser, 2007). We additionally apply scaling factors (e.g., 0.5 and 2.0) to LNOx emissions, producing LNOx at the lower and upper limits of the estimated range for sensitivity simulations described below. "

Line 249ff: "The meridional curve" needed some clarifications here, e.g., selected sites

within a latitude band to get the mean CH4 at 5 different latitude bands or something like that.

Reply: We have included detailed description in Section 2.2 in the revised manuscript as below:

"The global estimates are based on spatial and temporal smoothing of CH4 measurements from 45 surface marine boundary layer (MBL) sites. Locations of MBL sites are shown in Figure S1, and information for each MBL site is listed in Table S1 in the Supplement. First, the average trend and seasonal cycle are approximated for each sampling site by fitting a second-order polynomial and four harmonics to the data. We characterize deviations from this average behaviour by transforming the residuals to frequency domain, then multiplying by a low pass filter (Thoning et al., 1989; Thoning, 2019). Zonal and global averages are determined by extracting values at synchronized times steps from the smoothed fits to the data, then fitting another curve as a function of latitude (Tans et al., 1989). We divide these fits into sine (latitude) = 0.05 intervals, which define a matrix of zonally averaged CH4 as a function of time and latitude."

Line 296: Sometimes the sites like Key Biscayne are sampled by moving the model grids to the ocean side. You might check that out.

Reply: We thank the reviewer for the suggestion. Moving the model grid to the ocean side reduces the model bias. Similar issue also exists at Mace Head site.

Line 315ff: The tropical bias in all HIPPO is a bit strange! Not OH but transport (or emissions)? I am suspecting this because the bias due to OH would appear at all altitudes (timescale âĹij1yr), because the bias is in the lower troposphere, if the vertical transport is slow, you would find more CH4 is accumulated in the lower troposphere (timescale$\sim$week)

Reply: We agree with the reviewer that the bias could be due to transport or

emissions, which we already mentioned in the manuscript at lines 251-253. Also, OH is much higher over tropics than higher latitudes. It is possible that OH over tropics are overestimated in the model. Unfortunately, without observations, we are not able to rule out any possibilities.

Line 326: do you run CH3CCl3 SF6?, say within the CCMI framework?

Reply: We do not run with CH3CCl3 SF6.

Line 346ff: suggesting too much emissions in the NH, where most of Anthro emissions are...May be you can test this better by site-level comparisons.

Reply: We have updated all the plots in the revised manuscript due to a bug fix in the scripts for model evaluation. As shown in Figure 4 in the main text, the model is able to capture methane trend very well over high latitudes, with R = 1.0 and RMSE < 10 ppb in the Northern Hemisphere and RMSE $\sim$ 11 ppb in the Southern Hemisphere. For site-level comparisons, which are shown in Figures S4-5, over Northern Hemisphere, the model is able to reproduce methane DMFs at most of sites with RMSE < 25 ppb and R >0.9 although there are some overestimations at certain sites possibly due to overestimations in the local sources. Site-level comparisons are presented at lines 298-319 in the revised manuscript.

Line 353ff: I cannot find this 1 year mismatch (please be clear), instead I find a persistent offset during 1984-1991 (how the major and minor ticks marked in Fig. 5; the labeled ticks only should be major?

Reply: We have updated all the plots in the revised manuscript due to a bug fix in the scripts for model evaluation. As shown in Figure 4, the model fails to reproduce methane growth rates during 1984-1991, especially over tropics. This could be due in part to the fewer available observations used for emission optimization during this time period. The plots of methane growth rates are also updated with same major and minor ticks as shown for methane trend plots in

Figure 4.

Line 369: How can you say that? I thought your optimization was not good for this period, because the number of observation sites may not have covered the global reasonably well. I mean biased high toward the NH. Could you check how many SH sites you have data before 1988.

Reply: We agree with the reviewer that we have fewer observations available before 1988 used for emission optimization. Over Southern Hemisphere, there are only six sites (i.e., SPO, HBA, PSA, CGO, ASC, and SMO) as shown in Figure S5 that at least have one-year data available before 1988, which are much fewer than the number of sites over Northern Hemisphere.

Line 374: Most likely due to an overestimation of China emissions (e.g., Saeki and Patra, GOSL, 2017, and references therein) (regional inversion is needed for adjusting such regional emission biases)

Reply: We thank the reviewer for the references. We have included them in the revised manuscript as below:

"The overestimates are likely due to overestimation of emissions over Southeast Asia (e.g., Saeki and Patra, 2017, Patra et al., 2016, and Thompson et al., 2015), which could affect these remote sites through transport."

Line 378: "...which is also a remote site" and remote from China emissions

Reply: We agree with the reviewer's comment. This again suggests an overestimation in the emissions over Southeast Asia. We have included this in the revised manuscript as below:

"However, the model predicts surface methane DMF relatively well at Ascension Island (ASC, 8oS, 14.4oW, 85 m), which is also a remote site without impacts from East Asia."

Lines 394ff: I am not very sure if the comparison with GOSAT/SCIA are adding any values to this work. Better be kept aside for a full paper, unless the reasons for the mean offsets are figured out and discussed. For instance you could compare your results with the ACE-FTS data to find out if there is any bias in the stratospheric CH4 as there is no significant offsets in the tropospheric CH4 is seen in comparison with surface data and HIPPO.

Reply: We have moved the model evaluation against satellite observations to the Supplement.

Line 426ff: The emission increase in the 1990s is apparently linked to OH increase in AM; which sector can provide this extra emissions. I think this result is very different from what I have seen in the literature, and thus needing some explanation. Surprisingly, the emission increase rate in the 1990s is greater than the recent regrowth period.

Reply: In S0Wopt, the total emission growth during the 1980s is mainly from emission growth from agriculture (0.7 Tg yr-1), energy (0.3 Tg yr-1), waste (1.0 Tg yr-1), and wetland (1.8 Tg yr-1), while the total emission growth during the 1990s is mainly from waste (0.8 Tg yr-1) and wetland (3.7 Tg yr-1). Therefore, these extra emissions in the 1990s are mainly from wetland in S0Wopt scenario. In S0Aopt, the total emission growth during the 1980s is mainly from emission growth from agriculture (1.4 Tg yr-1), energy (0.9 Tg yr-1), and waste (1.4 Tg yr-1), while the total emission growth during the 1990s is mainly from agriculture (1.3 Tg yr-1), energy (1.1 Tg yr-1), waste (1.6 Tg yr-1), and biomass burning (0.4 Tg yr-1). Therefore, these extra emissions in the 1990s are mainly from energy, waste, and biomass burning sectors in S0Aopt scenario. The emission increase rate in the 1990s is greater than the recent growth period because OH decreases in the recent growth period. When we optimize emissions, we consider the impacts of OH trends. In the recent growth period, OH is decreasing, and therefore methane lifetime increases. This amplifies the responses of methane concen-

trations to the changes in the emissions. Therefore, a smaller increase in the emissions during this period can lead to larger increases in methane concentrations compared to that in the 1990s.

Lines 484ff: The discussions using Fig 9-11 aren't that interesting as presented. I would recommend the authors to move these plots to the supplement or show 1-2 panels in the main text; for example all the 4 panels in Fig 9 10 are essentially showing very similar distributions. The S0Aopt and S0Wopt are also showing similar behaviour. This is mainly because the emission (E)-a priori emissions are the same in both the simulations, and the correction emissions Del-E following Anthropogenic or Wetland emission patterns only play minor role. I am actually curious if you could use some of the continental sites, e.g., NWR, LEF, SGP or TAP, and use the model-measurement comparisons to say whether the S0Aopt or S0Wopt are more realistic.

Reply: We thank the reviewer for these suggestions. We have moved Figures 9-11 to Figures S7-10 in the Supplement. We also include site-level comparisons in the revised manuscript at lines 286-309. Despite the differences in the relative contributions of individual sources in S0Aopt and S0Wopt, due to the mixed-source effect and transport, the performance by S0Wopt and S0Aopt are very similar at most of sites. It is difficult to tell which one is more realistic in general. However, we do see general higher CH4 concentrations in S0Aopt than S0Wopt over the Northern Hemisphere. As shown in the Supplement Figure S4-5, over tropics, S0Wopt is in general better with smaller biases at KUM, POCN20, POCN25, and MID, which suggests overestimations in the Southeast Asian emissions. Also, anthropogenic emissions may be underestimated in S0Wopt at WLG and NWR and overestimated at TAP site based on the comparisons of S0Aopt and S0Wopt, whereas wetland emissions may be overestimated in S0Wopt at LEF site.

Line 519: Such high correlations are a bit surprising, if I see the lines in Fig. 12. For example AGR show -ve trend, yet show positive correlation. How is that possible?

Reply: The high correlation is mainly for periods of 1983-1998 and 2007-2014. In Figure 12 (now Figure 7 in the revised manuscript), the negative trend for AGR during 1999-2006 is very weak (i.e., -0.2 ppb/yr), almost no significant trend. The correlation here is more dominated by the interannual variability than linear trend. The positive correlation therefore suggests the interannual variability of CH4AGR agrees with that of total CH4.

Line 633: This is similar to the essential conclusion in some other publications as well, where ENE and Animals were made responsible for the post-2006 CH4 growth rate. I guess it is extremely difficult to separate emissions from Animals and Wetlands by 13C signature in CH4.

Reply: We agree with the reviewer's comments that rely on methane 13C signatures only is not able to distinguish emissions from animals and wetlands.

Lines 638ff: I am curious if inconsistency between the tropospheric OH and CH4-loss by OH are arising from the spin-up. Did you spun-up the simulations using different OH from the 1970s?

Reply: In our spin-up simulations, we drive the model with 1979 emissions for 50 spin-up years. During spin-up, OH changes every year based on the simulated chemistry. We used the spun-up atmospheric conditions for all the production runs, including low-OH and high-OH cases. In other words, we use the same initial conditions for low and high OH cases. As OH is short-lived species, the initial condition of OH has little impact on OH concentrations and trends. Based on our model tests, we find a very close relationship between tropospheric OH and lighting NOx. The changes in CH4-OH loss are not only affected by changes in OH but also CH4. Decreases in OH levels during 2008-2015 does necessary lead to decreases in CH4-OH loss as CH4 is increasing during this time period.

Please also note the supplement to this comment:

https://www.atmos-chem-phys-discuss.net/acp-2019-529/acp-2019-529-AC1-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-529, 2019.