Evolution of the global methane budget over 1980-2017 using the GFDL-AM4

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**Abstract**

Changes in atmospheric methane abundance have implications for both chemistry and climate as methane is both a strong greenhouse gas and an important precursor for tropospheric ozone. A better understanding of the drivers of trends and variability in methane abundance over the recent past is therefore critical for building confidence in predictions of future methane levels. In this work, the representation of methane is improved in the atmospheric chemistry model of ESM4 (AM4) and total methane emissions are optimized (with an annual mean of 576±32 Tg yr-1) based on methane mass balance to match surface observations for 1980-2017. The simulations with optimized global emissions are in general able to capture observed global methane trend and variability, seasonal cycle, and the latitudinal gradient. Simulations with different emission adjustments suggest increases in methane sources (mainly from energy and waste sectors) balanced by the increases in methane sinks (mainly due to increases in OH levels) leading to methane stabilization (with an imbalance of 6 Tg yr-1) during 1999-2006, and increases in methane sources combined with little change in sinks (despite small decreases in OH levels) during 2007-2012 leading to renewed methane growth (with an imbalance of 14 Tg yr-1 for 2007-2017). Compared to 1999-2006, both methane emissions and sinks are greater (by 31 Tg yr-1 and 22 Tg yr-1) during 2007-2017. The results also indicate the energy sector is more likely a major contributor to the methane renewed growth than wetland after 2006 as increases in wetland emissions alone are not able to explain the renewed methane growth under constant anthropogenic emissions. In addition, a significant increase in wetland emissions along with decreases in anthropogenic emissions starting in 2006 are required compared to the stabilization period (i.e., 1999-2006) for wetland emissions to drive renewed growth in methane, however, is a less likely scenario. Simulations with different OH levels indicate 1% change in OH levels could lead to an annual mean of ~ 4 Tg yr-1 difference in the optimized emissions and 0.08 year difference in the estimated tropospheric methane lifetime. Continued increases in methane emissions along with decreases in tropospheric OH concentrations during 2008-2015 prolong methane lifetime and therefore amplify methane’s response to emission changes. Uncertainties still exist in adjusting individual sources and regional emissions.

Keywords:

Atmospheric Methane, Methane Trend and Variability, Methane Budget, GFDL-AM4

**1 Introduction**

Atmospheric methane (CH4) is the second most important anthropogenic greenhouse gas with a global warming potential 28-34 times that of carbon dioxide (CO2) over a 100-year time horizon (Myhre et al., 2013). Methane is also a precursor for tropospheric ozone (O3), both an air pollutant and greenhouse gas, influencing its background levels (Fiore et al., 2002). Controlling methane has been shown to be a win-win for both climate and air quality (Shindell et al., 2012). From a preindustrial level of 722±25 ppb (Etheridge et al., 1998; Dlugokencky et al., 2005), methane has increased by a factor of ~2.5 to a value of 1850±1 ppb in 2017 (Dlugokencky et al., 2018) mostly due to anthropogenic activities (Dlugokencky et al., 2011). The global network of surface observations over the past 3-4 decades indicate that methane went through a period of rapid growth from the 1980s to 1990s, nearly stabilized from 1999 to 2006, and a renewed rapid growth until the present. Studies of drivers of observed changes in methane trends and variability have focused on the contributions from changes in methane sources and sinks. Here, we apply the next generation NOAA Geophysical Fluid Dynamics Laboratory chemistry-climate model, GFDL-AM4 (Zhao et al., 2018a, b) with improved representation of methane to explore the contribution of methane sources and sinks on its observed trends and variability.

The atmospheric methane budget is governed by its sources and sinks. Methane is emitted into the atmosphere from both anthropogenic activities (e.g., agriculture, energy, industry, transportation, waste management, and biomass burning), and natural processes (e.g., wetland, termites, oceanic and geological processes, and volcanoes), and is removed from the atmosphere mainly by reaction with hydroxyl radical (OH) in the troposphere (dominant), with smaller destruction by excited atomic oxygen (O1D) and atomic chlorine (Cl) in the stratosphere, and uptake by soils. Measurements of the global distribution of surface methane beginning in 1983 have revealed three major periods of methane growth: relatively larger increase prior to 1999 with the atmospheric methane growth rate of about 12 ppb yr-1 during 1984-1991 and 5 ppb yr-1 during 1992-1998 (Nisbet et al., 2014; Dlugokencky et al., 2018); stabilization during 1999-2006 with the growth rate of 0.7±0.6 ppb yr-1 (Dlugokencky et al., 2018); renewed growth from 2007 to 2013, with the growth rate of 5.7±1.2 ppb yr-1 that reached 12.6 ± 0.5 ppb yr-1 in 2014 and 10.0 ± 0.7 ppb yr-1 in 2015 (Nisbet et al., 2016; Dlugokencky et al., 2018).

The investigation on the drivers for the observed methane trend and interannual variability has mainly focused on the changes in global methane budget. While anthropogenic activities have been widely considered to largely contribute to the long-term methane increase since pre-industrial (Dlugokencky et al., 2011), there is no consensus on the drivers for the methane stabilization during 1999-2006 and renewed growth since 2007 in the scientific community. Previous studies have attributed the stabilization during 1999-2006 to the combined effects of increased anthropogenic emissions with decreased wetland emissions (Bousquet et al., 2006), decreased fossil fuel emissions (Dlugokencky et al., 2003; Simpson et al., 2012; Schaefer et al., 2016) or rice paddies emissions (Kai et al., 2011), stable emissions from microbial and fossil fuel sources (Levin et al., 2012), or variations of methane sinks (Rigby et al., 2008; Montzka et al., 2011; Schaefer et al., 2016). The observed renewed growth since 2007 has been explained on the basis of increases in wetland emissions (Dlugokencky et al., 2009; Bousquet et al., 2011; Nisbet et al., 2016), increases in agricultural emissions (Schaefer et al., 2016), increases in fossil fuel emissions (Rice et al., 2016; Worden et al., 2017), or decreases in sources compensated by decreases in sinks due to OH levels (Turner et al., 2017; Rigby et al, 2017). These different explanations reflect limitations in our understanding of recent changes in the methane and its budget.

Previous work has generally combined observations of methane and its δ13C with isotopic source signatures weighted by their emissions with inverse models (top-down), process-based models (bottom-up), or box models to estimate methane emissions and sinks and their variability (Bousquet et al., 2006; Monteil et al., 2011; Rigby et al., 2012; Bloom et al., 2012; Kirschke et al., 2013; Ghosh et al., 2015; Schwietzke et al., 2016; Schaefer et al., 2016; Nisbet et al., 2014, 2016; Dalsøren et al., 2016; Turner et al., 2017; Rigby et al., 2017). Inverse models use observations to derive emissions but usually prescribe climatological OH, O(1D), and Cl levels or loss rates (e.g., Rice et al., 2016; Tsuruta et al., 2017). Box-models, on other hand, use methane observations together with those of other proxy chemicals (e.g., 13C/12C ratio, ethane, carbon monoxide, methyl chloroform) to provide information on the global methane budget (e.g., Schaefer et al., 2016; Turner et al., 2017) but lack information on spatial variability or regional characteristics. With process-based models (e.g. wetlands) and inventories representing different source types (e.g. fossil fuel emissions) to drive chemistry transport models, the bottom-up approach is able to estimate the methane budget for all individual sources and sinks. However, without observational constraints, there is a potential bias in the total methane emissions derived from a combination of independent bottom-up estimates (Saunois et al., 2016).

Bottom-up global Earth System Models (ESMs) that realistically simulate the physical, chemical, and biogeochemical processes as well as interactions and feedbacks among these processes are useful tools to characterize the global methane cycle, and quantify the global methane budget and impacts on composition and climate. Dalsøren et al. (2016) investigated the atmospheric methane evolution by driving a chemistry transport model with bottom-up emissions. While their model results are able to match the observed time evolution of methane, without emission adjustments, surface methane is largely underpredicted in their study. Ghosh et al. (2015) optimized bottom-up emission data to investigate methane trends, however, OH trends and interannual variability were not considered in their chemistry transport model. Here, we apply the full chemistry version of the Geophysical Fluid Dynamics Laboratory (GFDL) new-generation Atmospheric Model, version 4 (AM4, Zhao et al., 2018a, b) to investigate the evolution of methane over 1980-2014. Our main objectives are toimprove the representation of methane in GFDL-AM4, comprehensively evaluate the model performance of methane predictions with improved representation of methane budget, and investigate possible reasons for the methane trends and variability. This paper is structured as follows: Section 2 describes the modeling approach, emission inventories, and observations used for model evaluation. Results of the model evaluation, global methane budget analysis, and model sensitivities are presented in Section 3. Finally, Section 4 summarizes the results and discusses the implication of these results.

**2 Methodology and data**

**2.1 Model description and initialization**

We use the full chemistry version of the next generation NOAA Geophysical Fluid Dynamics Laboratory chemistry-climate model, GFDL-AM4 (Zhao et al., 2018a, b). Detailed description of the physics and dynamics in AM4 is provided by Zhao et al. (2018a, b). AM4 with full interactive chemistry used in this is work is described by Schnell et al. (2018). In its standard form, this model setup consists of a cubed sphere finite-volume dynamical core with a horizontal resolution of ~100 km with 49 vertical levels extending from the surface up to ~80 km. The model’s lowermost level is approximately 30 m thick. The chemistry and aerosol physics in this model have been updated from the previous version of (GFDL-AM3; Naik et al., 2013) as described by Mao et al. (2013a, b) and Paulot et al. (2016). There are 102 total advected gas tracers and 18 aerosol tracers, 44 photolysis reactions, and 205 chemical reactions included in the chemical mechanism in this version of AM4 with full interactive tropospheric and stratospheric chemistry.

The standard AM4 configuration uses global annual mean methane concentrations as lower boundary conditions to simulate the atmospheric distribution of methane. Although the model simulates reasonable global mean methane abundances, large biases exist in the simulated latitudinal distribution and seasonal cycle. This modeling framework also does not allow for the full characterization of the drivers of methane trends and variability. To overcome this issue, we updated AM4 to be driven by methane emissions. Table 1 provides information on the emission datasets used in this work. Surface emissions from anthropogenic sources, including agriculture (AGR), energy production (ENE), industry (IND), road transportation (TRA), residential, commercial, and other sectors (RCO), waste (WST), and international shipping (SHP), are from the Community Emissions Data System (CEDS, version 2017-05-18, Hoesly et al., 2018) developed in support of the Coupled Model Intercomparison Project Phase 6 (CMIP6) for 1980-2014, and from a middle-of-the-road scenario of Shared Socioeconomic Pathways targeting a forcing level of 4.5 W m-2 (SSP2-4.5) developed in support of the ScenarioMIP experiment within CMIP6 (Gidden et al., 2018) for 2015-2017. Biomass burning (BMB) emissions for 1980-2014 are from van Marle et al. (2017) and for 2015-2017, from SSP2-4.5, and are vertically distributed over seven ecosystem-dependent altitude levels between the surface and 6 km above the surface, following the methodology of Dentener et al. (2006). Anthropogenic and biomass burning emissions are monthly gridded emissions with seasonal and annual variabilities. Natural emissions include wetland (WET) emissions from the WetCHARTs version 1.0 inventory (Bloom et al., 2017), ocean (OCN) emissions from Brasseur et al. (1998) and near-shore methane fluxes from Lambert and Schmidt (1993) and Patra et al. (2011), termites (TMI) from Fung et al. (1991), and mud volcanoes (VOL) from Etiope and Milkov (2004) and Patra et al. (2011). Wetland emissions and ocean emissions are based on climatological monthly mean without interannual variability. The remaining natural emissions are based on a climatological annual mean (repeated every month without seasonal variability). Trends in the total emissions and emissions from major sectors from this compilation over 1980 to 2017 are shown in Figure 1. Trends in total emissions are primarily driven by trends in ENE, AGR and WST emissions while BMB emissions contribute to interannual variability in this compilation. Anthropogenic and biomass burning emissions of other short-lived species also follow CEDS and SSP2-4.5 inventories. Natural emissions of other short-lived species are from Naik et al. (2013). Biogenic isoprene emissions are calculated interactively based on Guenther et al. (2006).

The methane sinks considered in AM4 include oxidation by OH radicals, Cl, and O(1D), and dry deposition. Since the model does not represent tropospheric halogen chemistry, it does not consider removal of methane by Cl in the troposphere, which has been shown to be extremely minor (Gromov et al., 2018). The dry deposition flux of methane is calculated based on a monthly climatology of deposition velocity (Horowitz et al, 2003) calculated by a resistance-in-series scheme (Wesely, 1989; Hess et al., 2000). This is used to mimic methane loss by soil uptake which accounts for about 5% of total methane sink (Kirschke et al., 2013; Saunois et al., 2016).

In this work, we included 12 additional methane source tagged tracers in the chemical mechanism in the AM4 to attribute methane sources from agriculture, energy, industry, transportation, residents, waste, shipping, biomass burning, ocean, wetland, termites, and mud volcanoes (i.e., CH4AGR, CH4ENE, CH4IND, CH4TRA, CH4RCO, CH4WST, CH4SHP, CH4BMB, CH4OCN, CH4WET, CH4TMI, and CH4VOL). The tracers are emitted from corresponding sources, and undergo the same chemical pathways and dynamics as the original CH4 tracer. For analysis, we combine CH4IND, CH4TRA, CH4RCO, and CH4SHP as other anthropogenic tracers (i.e., CH4OAT), and combine CH4OCN, CH4TMI, and CH4VOL as other natural tracers (i.e., CH4ONA).

Initially the model was spun up in a 50-year run with repetitive 1979 emissions until stable atmospheric burdens of methane and its source tagged tracers were obtained. After the spin-up, several sets of simulations were conducted for 1980-2017 to quantify the methane budget and investigate the impacts from methane sources and sinks (see Section 2.2). All the model simulations are forced with interannually-varying sea surface temperatures and sea ice from Taylor et al. (2000) prepared in support of the CMIP6 Atmospheric Model Intercomparison Project (AMIP) simulations, but the horizontal winds are nudged to the National Centers for Environmental Prediction (NCEP) Global Forecast System (GFS) reanalysis using a pressure-dependent nudging technique as implemented in AM3 simulations (Lin et al., 2012).

**2.2 Simulation Design**

We conduct several sets of hindcast simulations for 1980-2017 as listed in Table 2 to quantify methane budget and investigate the contributions of sources and sinks to methane trend and variability. Model simulation with the initial methane emissions inventory (Einit) described in Section 2.1 was found to largely underpredict the methane burden. Assuming that this mismatch is due to an imbalance in the simulated methane budget, we can either increase methane sources or decrease methane sinks to match the observations. We perform several optimization simulations that explore sensitivity to uncertainties in either methane emissions or OH levels, the dominant sink of the methane. Because OH trends and variability depend on a number of factors, including temperature, water vapor, O3, and emissions of nitrogen oxide (NO*x*), carbon monoxide (CO), and volatile organic compounds (VOCs), it is not straightforward to perturb OH. Previous work has shown that OH levels are highly correlated with NO*x* from lightning (Fiore et al., 2006; Murray et al., 2013). Therefore, we apply a scaling factor to lightning NOx (LNOx) emissions to indirectly adjust OH levels without influencing its variability. The LNOx emissions are calculated interactively based on Horowitz et al. (2003) as a function of subgrid convection parameterized in the model. The climatological mean LNOx emission simulated by AM4 is about 3.6 TgN yr-1, which is within the range of 2-8 TgN yr-1 estimated by previous studies (e.g., Schumann and Huntrieser, 2007). Here, we have tested the sensitivity of methane predictions to three assumptions : 1) OH levels simulated by the standard AM4 (referred as “S0”); 2) low OH levels via applying a scaling factor of 0.5 to the default LNOx emission calculations (referred as “S1”); 3) high OH levels via applying a factor of 2 to the default LNOx emission calculation (referred as “S2”). For each OH option, we begin with initial methane emissions and then optimize global total emissions to match simulated methane with surface observations. Different OH levels could lead to different estimations of the optimized total emissions, which provides a possible range for total methane emission estimation.

We apply a simple mass balance approach to optimize global total methane emissions following the methodology of Ghosh et al. (2015). The emission adjustments for each year are therefore the differences between the optimized global total emissions and the initial global total emissions (i.e., ΔE = Eopt – Einit). We do not optimize emissions for each grid cell as done in inverse modeling framework (Houweling et al., 2017). Instead, we uniformly apply the adjusted emission ratios (i.e., Eopt/Einit) globally for each year to get the spatial and temporal distribution of the optimized emissions. We assume that the spatial distribution of methane emissions from the initial emission inventories are the best available information we have. During the emission optimization, we do not optimize the spatial distribution but the global totals and the temporal variability. Considering the large uncertainties in the anthropogenic and wetland emissions, we perform two simulations with ΔE added onto anthropogenic sources, including biomass burning sector (referred to as “Aopt”) and with ΔE added onto wetland sector only (referred to as “Wopt”) for S0 scenario. The purpose of conducting these simulations is to investigate the impact of optimizing emissions from different sectors on methane predictions. For the Aopt case, ΔE is partitioned into eight anthropogenic sectors (i.e., AGR, ENE, IND, TRA, RCO, WST, SHP, and BMB) by applying the ratio of ΔE to total anthropogenic emissions to each of the total eight anthropogenic sectors, which means the fractions of individual sources to total sources are kept the same before and after the emission optimization. For the Wopt case, ΔE is added onto wetland emissions only. For S1 and S2 scenarios, we apply ΔE to wetland sector only. The total Eopt emissions are the same for both Aopt and Wopt cases. Time series of methane optimized total emissions and emissions from major sectors from S0Aopt and S0Wopt over the 1980 to 2017 period are shown in Figure S2 in the Supplement.

**2.3 Observations**

We evaluate the simulated surface methane dry-air mole fraction (DMF) against a suite of ground-based and aircraft observations, and satellite retrievals of column-averaged CH4 to thoroughly evaluate the model simulated spatial and temporal distribution of methane. To evaluate surface CH4, we use measurements from a globally distributed network of air sampling sites maintained by the Global Monitoring Division (GMD) of the Earth System Research Laboratory at the National Oceanic and Atmospheric Administration (NOAA) (Dlugokencky et al., 2018). The global estimates derived from surface measurements are based on a number of sites at remote marine sea level locations with well-mixed marine boundary layer (MBL) to represent background methane. The locations of MBL sites are shown in Figure S1 and the information for each MBL site is listed in Table S1 in the Supplement. The fast Fourier transform and low-pass filters (Thoning et al., 1989) are applied to the monthly mean methane DMFs to approximate the long-term trend and average seasonal cycle at each MBL site. A meridional curve (Tans et al., 1989) was fitted through these site values to get the latitudinal distribution of methane. The same sampling and processing approach (Thoning et al., 1989; Tans et al., 1989) is applied to the simulated monthly mean methane DMF to calculate global and zonal averages to facilitate consistent model-observation comparison. Besides the comparison with global estimates from MBL sites, we also evaluate the model performance at various GMD sites to investigate the contributions from local sources. For site specific evaluation, we sample the model grid cell at the location of the corresponding site and at the model layer with height closest to the altitude of the corresponding site.

Due to the sparseness of the ground-based observational sites, especially over continental regions, we also evaluate simulated methane against satellite retrievals to reveal information on regional characteristics. Total column-averaged methane DMFs are evaluated against satellite retrievals from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) instrument on board the European Space Agency’s environmental research satellite ENVISAT (Frankenberg et al., 2011) for January 2003 to April 2012 and the Thermal And Near Infrared Sensor for carbon Observations – Fourier Transform Spectrometer (TANSO-FTS) instrument on board the Japanese Greenhouse gases Observing SATellite (GOSAT) (Kuze et al., 2016) for April 2009 to December 2016. We compare monthly mean satellite retrievals with simulated monthly mean methane. Retrieval-specific averaging kernels are also applied to simulated monthly mean methane to calculate simulated column-averaged methane DMF.

To investigate background tropospheric methane variability, we compare the simulated methane vertical profiles with aircraft measurements from HIPPO– the High-performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole observation (HIPPO) campaigns from January 2009 to September 2011 (Wofsy et al., 2012). A total of 787 profiles were flown during the 5 campaigns with continuous profiling between approximately 150 m and 8500 m altitudes, but also including many profiles of up to 14 km altitude. For each HIPPO mission, we sample the model consistently in the same space as the observations and average the model for the corresponding months that the campaign occurred to create climatological monthly means.

**3 Results and discussions**

**3.1 Model evaluation**

The detailed model evaluation for S0Aopt and S0Wopt are discussed below. We first evaluate the mean climatological spatial distribution and seasonal variability simulated by the model and then evaluate the simulated trends and variability.

**3.1.1 Climatological evaluation**

Figures 2a and b show the model bias and correlation coefficient of simulated climatological mean surface methane DMF against NOAA GMD surface observations (Dlugokencky et al., 2018) for the 1983-2017. The mean seasonal cycle at individual GMD sites is shown in Figure S3 in the Supplement. GMD sites with at least 20 years of observations are selected for model climatological evaluation. The information of these sites is shown in Table S2 in the Supplement. As shown in Figure 2a, simulations with optimization of either anthropogenic (S0Aopt) or wetland (S0Wopt) emissions are generally able to reproduce surface methane DMF with model biases within ±30 ppb at most of the sites. Both S0Wopt and S0Aopt simulate methane DMF relatively well over the Southern Hemisphere. Going from South to North, the low bias in methane DMF decreases and becomes a high bias over the tropics. Simulated methane in both S0Aopt and S0Wopt are moderately high biased in the tropical Pacific Ocean (by up to ~ 40 ppb), indicating possible overestimationof methane emissions over the tropics and possible underestimations in OH levels. Large positive biases occur at Key Biscayne (25.7 N, 80.2 W) for both S0Wopt and S0Aopt, mainly due to the overpredictions during summer and fall (see Figure S3 in the Supplement). Over middle and high latitudes of Northern Hemisphere, the simulated surface methane DMF are both low and high biased, possibly in part due to the uncertainties in the emissions representing local sources. As shown in Figure 2b, both S0Aopt and S0Wopt are able to capture the methane seasonal cycle at most sites (with a correlation coefficient (R) larger than 0.5 for about 80% of total sites). Both S0Aopt and S0Wopt are able to reproduce the methane seasonal cycle over the Southern Hemisphere. However, both S0Aopt and S0Wopt show poor performance in seasonal cycles over the tropical Pacific Ocean in the Southern Hemisphere, with R < 0.5 (e.g., POCS10 and POCS15 in Figure S3 in the Supplement) but show good performance in seasonal cycles over the tropical Pacific Ocean in the Northern Hemisphere, with R = 0.9 (e.g., POCN05, POCN10, and POCN15 in Figure S3 in the Supplement). Poor performance also exists at a few sites in middle and high Northern Hemisphere (e.g., AZR, UUM, LEF, MHD, and ICE shown in Figure S3 in the supplement), mainly due to overprediction of methane during summer. Uncertainties in the seasonal variations of methane emissions, OH predictions and long-range transport could lead to biases in predicting a seasonal cycle. In general, both S0Aopt and S0Wopt are able to capture the methane latitudinal gradient (e.g., R = 0.9). This suggests that the methane spatial distribution in the initial emissions are overall reasonable despite uncertainties in representing local sources.

To investigate background tropospheric methane variability, Figure 3 shows the simulated vertical distribution of methane minus that observed during a HIPPO campaign for the S0Aopt and S0Wopt simulations. S0Aopt and S0Wopt simulations predict very similar methane profiles. Both S0Aopt and S0Wopt predict methane profiles very well over Southern Hemisphere. Compared to HIPPO measurements, methane in both simulations is consistently biased high over the tropical Pacific Ocean (by up to ~ 50 ppb) from the surface to 700 mb during all HIPPO missions. But the biases decrease with altitude and decrease with latitude except for summer. In the Northern Hemisphere, both S0Wopt and S0Aopt simulations capture the observed methane near the surface to 700 mb, but are biased low except in summer when they are biased high, especially at mid-latitudes. Middle latitude background methane is affected by both high-latitude and low-latitude air masses on synoptic scales. Biases over these regions could result from many processes (e.g., overestimation in the summer emissions, underpredictions of OH levels, and model transport). In general, the relative differences between the simulated methane profiles and HIPPO measurements are within 2% over most regions, demonstrating the capability of the improved GFDL-AM4 for tropospheric methane predictions.

**3.1.2 Trend evaluation**

As described in Section 2.3, we applied the fast Fourier transform and low-pass filters (Thoning et al., 1989) to estimate the long-term trend and growth rates discussed below. The comparisons of simulated global mean background surface methane trends and growth rates to NOAA-GMD observations are shown in Figures 4 and 5. Both S0Wopt and S0Aopt predict similar global mean surface methane DMF, trend, and growth rate since the global methane budget (emissions and sinks) are the same in the two simulations. S0Wopt and S0Aopt are also able to reproduce the global mean surface methane DMF (with root-mean-square-error (RMSE) = 8.3 ppb in S0Wopt and 8.9 ppb in S0Aopt) averaged over t1983-2017 and capture the methane trend very well (with R = 1.0 in both S0Wopt and S0Aopt), especially over the Southern Hemisphere. In general, the RMSE for S0Wopt is lower than that for S0Aopt, except over the low Southern Hemisphere. The major discrepancies in the surface methane DMF between model simulations and observations are mainly in the low Northern Hemisphere (0-30o N), especially the tropics (Figure 4d), where the RMSE is greater than 20 ppb. Over the high Northern Hemisphere, S0Aopt overpredicts background methane DMF by about 20-30 ppb during 1984-1998, whereas S0Wopt has better performance during this period. After 1998, S0Aopt can reproduce the maximum methane DMF very well while S0Wopt slightly underpredicts methane DMF by up to 10 ppb. The agreement between simulated and observed global methane trend demonstrates the confidence in the optimized methane emission trends used in this work.

As shown in Figure 5, both simulations are in general able to able to reproduce the observed global methane growth rate (with R = 0.8 in both S0Wopt and S0Aopt), despite a slight mismatch (~1-year) during 1997-2007. Global methane simulated by S0Aopt grows rapidly (annual mean ± standard deviation) during 1984-1991 (i.e., 13.5±2.1 ppb yr-1), slower growth during 1992-1998 (i.e., 4.8±2.6 ppb yr-1), a relative stabilization during 1999-2006 (i.e., 1.2±4.7 ppb yr-1), and a renewed growth during 2007-2017 (i.e., 6.1±2.8 ppb yr-1). The simulated global methane growth rates by S0Wopt show a similar trend (i.e., 13.5±2.1 ppb yr-1 during 1984-1991, 4.7±3.3 ppb yr-1 during 1992-1998, 1.3±4.8 ppb yr-1 during 1999-2006, and 6.1±2.8 ppb yr-1 during 2007-2017). The simulated growth rates during 1984-1991 are slightly higher than the NOAA-GMD estimates (i.e., 11.6±1.3 ppb yr-1), while the simulated growth rates during 1992-1998, 1999-2006, and 2007-2017 are within the ranges of NOAA-GMD estimates (i.e., 5.6±3.5 ppb yr-1, 0.7±3.1 ppb yr-1, 6.9±2.6 ppb yr-1). Over the tropics, both S0Aopt and S0Wopt overpredict methane growth rates (by about 5-10 ppb yr-1) during 1984-1990, but are able to reproduce methane growth rates relatively well afterwards. Agreement of the methane growth rate predictions is worse in the Northern Hemisphere than in the Southern Hemisphere, especially over the high Northern Hemisphere, where R is smaller than 0.5. Over 30-90oN, neither S0Aopt nor S0Wopt is able to reproduce methane growth rates during 1984-1989 and there is a slight mismatch (~1-2 years) in methane growth rates predictions afterwards. These biases indicate larger uncertainties in the methane emissions in the high Northern Hemisphere than other regions.

Comparisons of simulated surface methane DMF to NOAA-GMD observations are also conducted at individual sites shown in Figure S4 in the supplement. S0Aopt and S0Wopt simulations predict very similar methane DMF. Both simulations tend to be biased low over Southern Hemisphere sites, but the underpredictions decrease northward. The simulations are moderately high biased (with RMSEs up to ~ 40 ppb) over tropical regions (e.g., POCS15, POCS10, SMO, POCS05, POCN00, CHR, and POCN05). These sites are mainly remote sites and surface methane DMF represents background methane levels. However, the model predicts surface methane DMF relatively well at ASC (i.e., Ascension Island, 8oS, 14.4oW, 85 m), which is also a remote site. The altitude of ASC is much higher than other remote sites. This indicates that the model tends to overpredict background methane DMF at the surface but is able to capture background methane at higher levels. Moderate overpredictions also occur at SEY (i.e., Mahe Island, 4.7oS, 55.5oE), which could be affected by airmasses from polluted areas over the tropics and Northern Hemisphere. Over middle and high Northern Hemisphere, both S0Aopt and S0Wopt relatively well predict surface methane DMF at most of the sites, except at KEY (Key Biscayne, 25.7oN, 80.2oW), TAP (Tae-ahn Peninsula, 36.7oN, 126.1oW), LEF (Park Falls, 45.9oN, 113.7oW), and MHD (Mace Head, 53.3oN, 9.9oW). KEY and MHD are remote sites, whereas TAP and LEF are affected largely by local sources and model transport. The overpredictions could be in part due to the uncertainties in representing local emissions. On the other hand, both S0Wopt and S0Aopt are able to capture the monthly variation at most of the sites except at LEF, where R is 0.4 for S0Wopt and 0.5 for S0Aopt, respectively. In general, both S0Wopt and S0Aopt are able to reproduce the surface methane DMF and capture the trend at most sites (e.g., with R greater than 0.5 at 98% of total sites and with RMSE less than 30 ppb at 74% of total sites).

Unlike evaluation of global mean surface methane DMF, which is based on the observations derived from a number of sites representing the well-mixed MBL, the evaluation of global mean column-averaged methane DMF against satellite retrievals mainly covers continents, considering the impacts from polluted areas and the contributions from troposphere and stratosphere. The comparisons of simulated monthly mean column-averaged methane DMF to satellite retrievals (e.g., SCIAMACHY and GOSAT) are shown in Figure 6. The averaging kernels of SCIAMACHY and GOSAT are individually applied to the model to calculate column-averaged methane abundances. Both simulations are able to capture the monthly variation of methane with R greater than 0.9, but underestimate column-averaged methane, with RMSE of about 21 ppb and 29 ppb when compared to SCIAMACHY and GOSAT retrievals, respectively. The biases increase poleward in both SCIAMACHY and GOSAT comparisons; large uncertainty exists in the satellite retrievals over high latitudes (e.g., > 70o) due to high solar zenith angles and potential high cloud cover over. The underpredictions are mainly due to the uncertainties in the methane predictions in middle/upper troposphere and stratosphere (as shown in Figure 3). The differences in the column-averaged methane abundances between satellite retrievals and model simulations are within 2% over most regions except Polar regions where there are large uncertainties in the satellite retrievals. Both simulations are also able to capture the latitudinal distribution of the column-averaged methane DMF with R close to 1.

**3.2 Global methane budget**

Figure 7 shows time series of optimized total CH4 emissions, global sink, and global burden based on S0Wopt. Since S0Aopt and S0Wopt simulations are very close to each other for global totals, we only show the budget for S0Wopt. As depicted in Figure 7, the simulated global methane burden steadily increases from 1980 to 1992, with a burden growth rate of 39 Tg yr-1. During 1993-1998, the global methane burden growth slows down with a growth rate of 16 Tg yr-1. The growth rates simulated by the model agrees well with the observed growth rates during 1984-1997 as shown in Figure 5a. The simulated growth rate in global methane burden decreases to 4 Tg yr-1 during 1999-2006 while it increases to 16 Tg yr-1 during 2007-2017 and reaches over 20 Tg yr-1 during 2014-2016. The changes in the global burdens are due to the imbalance between methane sources and sinks. As shown in Figure 7, the optimized emissions in general increase during 1980-2017, with an annual mean of 576±32 Tg yr-1 (mean±standard deviation) and show much larger interannual variability during 1991-1993 and 1997-2000. Although there is an overall increasing trend in total global emissions, growth in annual mean emissions has increased from the 1980s (i.e., 3.9 Tg yr-1) to the 1990s (i.e., 4.4 Tg yr-1) and reached 7.0 Tg yr-1 during 1999-2006, but decreased to 2.3 Tg yr-1 during 2007-2017. The estimations of optimized emissions are based on the comparisons of simulated surface methane with surface observations. The uncertainties in the interannual variability of simulated OH levels and therefore methane sinks could lead to the uncertainties in the interannual variability of the optimized emissions. Also, since all the emissions are prescribed, the interannual variability of the optimized emissions also reflects the impacts from natural variability on methane emissions. The larger interannual variabilities during 1991-1993 and 1997-2000 are likely due to the strong El Nino events during 1991-1992 and 1997-1998.

Unlike methane emissions, the methane sink increases during 1980-2007, with relative stabilization during 2007-2014 but increases during 2015-2017. The annual mean methane sink during 1980-2017 is 560±44 Tg yr-1 (mean±standard deviation). The trends in methane sink are affected by the changes in both methane and OH levels (assuming that other sinks are minor). Figure 8 shows the tropospheric OH anomalies with respect to 1998-2007. An interesting finding is that AM4 predicts higher OH levels during 2007-2014 than 1998-2007 by 3.1%, whereas recent studies applying multispecies inversion with a box-model framework (e.g., Rigby et al., 2017; Turner et al. (2017) suggest a decline in OH levels after 2007. However, a recent study by Naus et al. (2018) found a shift to positive OH trend over 1994-2015 after applying bias corrections based on a 3-D CTM to a similar box model setup. In addition, OH levels simulated by AM4 decrease from 2013 to 2015 but increase again afterwards, leading to increases in methane sinks during 2015-2017. As shown in Figure 7, higher methane sources than sinks during 1980-1998 leads to the increases in methane burdens. A relative balance between methane sources and sinks during 1999-2006 leads to the methane stabilization. Compared to 1999-2006, both methane sources and sinks are higher during 2007-2017, but methane sources outweigh sinks after 2007 leading to renewed methane growth.

Table 3 provides a summary of decadal mean methane budget for 1980-2017. Compared to Kirschke et al. (2013) and Saunois et al. (2016), the total natural sources from the initial emission inventories (i.e., 203 Tg yr-1) are at the lower range of top-down estimates during this period, except for 1990s, when they are slightly higher than top-down estimates but still much lower than the bottom-up estimates. This is mainly because wetland emissions from the initial emission inventories are slightly higher than top-down estimates but much lower than bottom-up estimates during 1990s. The total anthropogenic sources from the initial emission inventories are overall within the range of top-down or bottom-up estimates, except for 1980-1989, when they are lower than the estimates in Kirschke et al. (2013) and Saunois et al. (2016). This is mainly due to lower emission estimates from agriculture and waste sectors in the CEDS inventory during this period. With the optimized global total emissions (i.e., ΔE proportionally added onto the initial anthropogenic sectors as in Aopt or ΔE added onto wetland sector only as in Wopt), the total sources used in this work and the total sinks estimated by AM4 are either in the range of top-down or bottom-up estimates. As a result, the imbalance between total sources and total sinks estimated in this work are overall within the range of estimates by previous studies although smaller imbalance than previous estimates exists for 2000s and afterwards. The atmospheric growth rates simulated by the model (sampled identically as for observations) are also comparable to the observed atmospheric growth rates.

**3.3 Sensitivity to sector optimization**

**3.3.1 Spatial distribution**

As described in Section 2.2, the emission optimization is conducted for anthropogenic sectors (i.e., S0Aopt) and wetland sector (i.e., S0Wopt). Global total methane emissions are the same for S0Aopt and S0Wopt, but there are different allocations for anthropogenic and wetland sectors and different spatial distributions as well, depending on the spatial distributions of the initial emission dataset. Here we analyze the sensitivity of sector optimization on spatial distribution of methane predictions. Figures 9 and 10 show the spatial distributions of the differences in the methane emissions and surface methane abundance between S0Aopt and S0Wopt during the four periods (i.e., 1980-1989, 1990-1999, 2000-2006, and 2007-2017). Surface methane is always lower in Aopt than Wopt in the tropics (e.g., 15o S-10o N) during the four periods. This is mainly due to much lower wetland emissions in S0Aopt than in S0Wopt (Figure 10), which dominates total emissions over these regions (e.g., tropical South America and Central Africa). There is not much difference in surface methane predictions over low and high Southern Hemisphere (e.g., 15-90o S) between the two simulations. This is mainly because larger anthropogenic emissions in S0Aopt compensate smaller wetland emissions and therefore the differences in the total emissions are very small, within 0.1 Tg yr-1 (Figure 10). Unlike Southern Hemisphere, surface methane concentrations are in general higher in S0Aopt than S0Wopt in the Northern Hemisphere, especially over Eastern U.S. and Eurasia, due to much higher anthropogenic emissions in S0Aopt. The lower surface methane values in S0Aopt over northern Canada are due to much lower wetland emissions in S0Aopt.

Figure 11 shows the methane growth rates simulated by Aopt and Wopt during the four time periods. Global mean methane growth rates simulated by Aopt and Wopt are very consistent during the four periods, with growth rates decreasing from 1980s to 1990s, stabilizing during 2000-2006, and increasing after 2007. During the 1980s and 1990s, methane growth rates in both S0Aopt and S0Wopt increase over most of the globe except a decrease over Russia, due to significant decreases in anthropogenic emissions (mainly from energy sector) in the former Soviet Union consistent with previous studies (Dlugokencky et al., 2011). During 2000-2006, methane growth rates increase significantly over East Asia in both S0Aopt and S0Wopt while they decrease over tropical South America and Central Africa in S0Wopt but not in Aopt. This is mainly due to decreases in wetland emissions in the S0Wopt case, while wetland emissions are constant for each year in Aopt case. After 2007, both Aopt and Wopt suggest large increases in methane growth rates over East Asia (mainly due to increases in anthropogenic emissions) by up to ~38 ppb yr-1 with smaller increases elsewhere (< 7 ppb yr-1) but noticeable increases over the Arctic ( > 7 ppb yr-1). This relatively large methane growth over the Arctic mainly occurs after 2014, possibly due to a sharp decrease in OH levels during 2014-2015.

As discussed in Sections 3.1 and 3.2, the similarity in S0Aopt and S0Wopt simulation results suggests that for 3-dimensional chemistry transport models, reasonable estimates of total global methane emissions are critical for global methane predictions despite the uncertainties in the spatial distribution of the emissions and in the estimates of individual sources, which are more important for regional methane predictions. At the same time, accurate estimates of individual sources are necessary to attribute methane trend and variability into individual sources.

**3.3.2 Source tagged tracers**

In this section, we apply Mann-Kendall (M-K) test to estimate the linear trend (different from long-term trend discussed in Section 3.1.2) of global mean source tagged tracers and total methane for 1983-1998, 1999-2006, and 2007-2017 to investigate possible drivers in total methane trends. Figure 12 compares the trends of source tagged tracers and total methane from S0Aopt and S0Wopt during 1983-1998, 1999-2006, and 2007-2017. As shown in Figure 12, both S0Aopt and S0Wopt are in general able to capture the methane trends during different time periods. For S0Aopt, globally, total methane show an increasing trend with linear increasing rate of 10.5 ppb yr-1 during 1983-1998, slightly overpredicting the increasing trend compared to observations (i.e., 8.8 ppb yr-1) but correlating very well with the observations (i.e., R = 1.0). All the anthropogenic tracers show increasing trends during 1983-1998 despite the increases in OH levels, with larger increasing trends by AGR (i.e., 3.6 ppb yr-1) and WST (i.e., 3.6 ppb yr-1) consistent with emission trends. Major anthropogenic tracers (e.g., CH4AGR, CH4ENE, and CH4WST) correlate very well with total methane, with R varying between 0.9 to 1.0 over this time period. Since wetland emissions and other natural emissions are constant every year, with the increases in OH levels during 1983-1998, both CH4WET and CH4ONA (i.e., CH4OCN+CH4TMI+CH4VOL) decrease, with a linear decreasing trend of -0.5 ppb yr-1 and -0.1 ppb yr-1 over this period. During 1999-2006, total methane has a small increasing trend of 1.3 ppb yr-1, still slightly overpredicting the increasing trend compared to observations (i.e., 0.6 ppb yr-1) but correlating relatively well with the observations (i.e., R = 0.8). During this time period, there are increasing trends in CH4ENE (i.e., 2.6 ppb yr-1) and CH4WST (i.e., 2.3 ppb yr-1) with slightly decreasing trends in CH4AGR (i.e., -0.1 ppb yr-1), CH4BMB (i.e., -0.9 ppb yr-1) and CH4OAT (i.e., CH4IND+CH4TRA+CH4RCO+CH4SHP, -0.5 ppb yr-1). Anthropogenic tracers such as CH4ENE and CH4WST correlate well with total methane, whereas CH4AGR show a poor correlation with total methane, and CH4BMB and CH4OAT show an anticorrelation with total methane over this time period. Similarly, with the increases in OH levels during 1999-2006, both CH4WET and CH4ONA decrease, with a linear decreasing trend of -1.8 ppb yr-1 and -0.4 ppb yr-1. During 2007-2017, total methane show a renewed increasing trend of 5.3 ppb yr-1, slightly underpredicting the increasing trend compared to observations (i.e., 6.0 ppb yr-1) but correlating relatively well with the observations (i.e., R = 1.0). During this time, CH4ENE show a large increasing trend (i.e., 5.8 ppb yr-1), dominating the total methane trend. Interestingly, although there is a slight decrease in OH levels after 2008, with both CH4WET and CH4ONA still show decreasing trends of -1.1 ppb yr-1 and -0.3 ppb yr-1 during 2007-2017. Also, all the natural tracers show an anticorrelation with total methane during this period. The results from S0Aopt suggest that globally, anthropogenic tracers dominate total methane trends during the entire simulation period. During the 1980s and 1990s, emissions from agriculture, energy, and waste sectors are the major contributors to the methane increase. During 1999-2006, where methane stabilizes, increases in methane sinks and methane sources take turn to dominate the trend for different tracers and therefore the imbalance between methane sinks and sources dominate the total methane trend. During 2007-2017, the energy sector is the major contributor to the methane renewed growth.

However, source tagged tracers behave slightly different in S0Wopt. For S0Wopt, globally, total methane show a similar increasing trend as S0Aopt (as discussed in section 3.1.2). All the anthropogenic tracers show increasing trends during 1983-1998 except CH4ENE (i.e., -0.3 ppb yr-1). Anthropogenic tracers (except CH4OAT) in general correlate well with total methane. CH4WET show a significant increasing trend during this period (i.e., 7.0 ppb yr-1) and correlate relatively well with total methane. During 1999-2006, anthropogenic tracers such as CH4ENE and CH4WST show increasing trends (i.e., 1.8 ppb yr-1 and 1.9 ppb yr-1) and correlate relatively well with total methane, whereas all other tracers show decreasing trends and are anticorrelated with total methane. During this time, our wetland tracer (i.e., CH4WET) shows a slightly decreasing trend (i.e., -0.6 ppb yr-1), mainly due to the slightly higher CH4WET sinks (i.e., 226 Tg yr-1) than sources (i.e., 223 Tg yr-1). During 2007-2017, anthropogenic tracers such as CH4AGR, CH4ENE, and CH4WST show significant increasing trends (i.e., 2.3 ppb yr-1, 6.9 ppb yr-1 , and 1.6 ppb yr-1) and correlate quite well with total methane whereas all other tracers except CH4OAT show decreasing trends and poor correlation with total methane. On the other hand, CH4WET shows a significant decreasing trend during this period (i.e., -4.6 ppb yr-1) and an anticorrelation with total methane. The decreasing trend of CH4WET is due to higher CH4WET sinks (i.e., 217 Tg yr-1) than sources (i.e., 206 Tg yr-1) during this period. In general, the results from S0Wopt suggest that globally, during 1983-1998, wetlands are the major contributor to the methane increase, which contradict previous studies that suggest anthropogenic emissions as the major contributors (e.g., Dlugokencky et al., 2011). During 1999-2006, when methane stabilizes, increases in methane emissions from energy and waste sectors dominate the increases in total methane sources as well as their tagged tracers (i.e., CH4ENE and CH4WST), whereas increases in methane sinks dominate all other tracers. Therefore, the imbalance between total methane sinks and sources dominate the total methane trend, which is also the case in S0Aopt during this time. During 2007-2017, energy is the major contributor to the methane renewed growth similar to that in S0Aopt.

As shown in Figures 7 and 8, OH levels show a slight decrease and methane sinks are relatively stable during 2007-2013 but large interannual variability exists during 2013-2017. Decreasing OH levels could lead to increases in methane lifetime and therefore methane buildup. Combined with increases in the emissions, methane starts to increase again during this period. However, it is difficult to separate the contributions from methane emissions and sinks as optimized methane emissions are based on methane mass balance (e.g., changes in the methane loss would act as a feedback on estimates of optimized total emissions). But, it is clear that the decrease in OH levels alone (for example if emissions are kept constant) would not be enough to reproduce the renewed growth. The question then is which emission sector(s) is(are) the major contributor(s) to the renewed growth over 2007 to 2017. Both S0Wopt and S0Aopt suggest that energy is the major sector contributing to renewed CH4 growth. However, both cases depend largely on the initial emission inventory. For example, S0Wopt does not influence the emission growth of other sectors in the initial emission inventory, which means if the emission growth of a certain sector in the initial emission inventory is overestimated or underestimated, it could have different implications.

Based on evidence from isotopic composition (δ13CH4), recent studies suggest increasing wetland emissions may be responsible for the renewed growth of methane (Dlugokencky et al., 2009; Nisbet et al., 2016). To test this in our modeling framework, we conducted another sensitivity simulation for 2006-2014, by repeating 2006 anthropogenic emissions for all the years but adjusting wetland emissions to ensure that the total methane emissions are the same as in S0Wopt (or S0Aopt), which would imply that the increases in methane emissions are only due to the increases in wetland emissions. This sensitivity simulation is referred to as “S0A06” and the trends for source tagged tracers and total methane are shown in Figure S5 in the supplement.

Interestingly, in S0A06, emissions from anthropogenic tracers still increase during 2007-2014, with the trend in CH4ENE dominating (i.e., trend = 3.6 ppb yr-1 and R = 1.0), whereas CH4WET shows a small decreasing trend (i.e., trend = -1.0 ppb yr-1 and R = -0.8). As OH levels slightly decrease during this time period, with constant emissions except wetland, it would expect possible increasing trends in all the source tagged tracers except CH4WET. In fact, CH4AGR, CH4ENE, CH4WST, and CH4BMB in S0A06 increase over 2007-2014 but with smaller increasing rates than those in S0Wopt (and S0Aopt) due to no emission growth for these tracers. On the other hand, the decreasing OH levels (Figure 8) would lead to less methane sink and therefore higher methane concentrations. But the methane loss is proportional to the product of OH levels and methane concentrations. Since contributions of CH4WET are much greater than other source tagged tracers, the loss of CH4WET are also much higher than other tracers. Higher CH4WET loss (i.e., 224 Tg yr-1) than CH4WET sources (i.e., 207 Tg yr-1) leads to a decreasing trend in CH4WET. Nevertheless, S0A06 results still suggest that the renewed growth during 2007-2014 is dominated by the increases of CH4ENE, which means OH trends play an important role in determining the increasing trend of total methane since emissions for the energy sector are kept constant in this sensitivity simulation. In addition, increases in wetland emissions alone are not able to drive increases in CH4WET as CH4WET sinks are equally important for determining the trend in CH4WET under constant anthropogenic emissions condition. S0A06 results also suggest that no matter how one allocates the emissions into individual sectors, the model is still able to capture the global methane trend as long as the global total methane emissions are well constrained.

We perform an additional sensitivity simulation to test the possibility of wetland emissions driving the methane trend during the period of renewed CH4 growth by combining the emissions of S0Aopt and S0Wopt as follows: S0Aopt emissions for 1980-2005 and S0Wopt emissions for 2006-2014. This simulation is referred to as “S0Comb” and the trends for source tagged tracers and total methane are shown in Figure S5 in the supplement. For 2007-2014, all anthropogenic tracers show decreasing trends except CH4ENE (i.e., 2.7 ppb yr-1) whereas CH4WET shows a significant increasing trend (i.e., 6.3 ppb yr-1) and dominates the total methane trend. This is mainly due to lower anthropogenic emissions during this period than previous periods and methane sinks of anthropogenic tracers start to take over their trends expect for CH4ENE. At the same time, significant higher wetland emissions during this period than previous periods dominate the increasing trend of CH4WET. Interestingly, even with the same wetland emissions in S0Wopt and S0Comb for 2006-2014, CH4WET shows different trends. This is mainly because the CH4WET GMFs at the beginning of 2006 are much lower in S0Comb than in S0Wopt. Therefore, CH4WET loss is much lower in S0Comb (i.e., 190 Tg yr-1) compared to S0Wopt (i.e., 220 Tg yr-1) over this time period, leading to increasing CH4WET trend in S0Comb but decreasing trend in S0Wopt. S0Comb results suggest the need for a significant increase in wetland emissions along with decreases in anthropogenic emissions starting in 2006 compared to the stabilization period for wetland emissions to drive renewed growth in methane. However, this is a less likely scenario as both top-down and bottom-up inventories indicate anthropogenic emissions increasing over the 2007-2014. A more likely scenario is that both anthropogenic and wetland emissions increase (e.g., higher during 2007-2014 than 1999-2006). However, in that case, the weight of the dominancy of wetland emissions on the total methane trend would decrease based on our analysis.

**3.4 Sensitivity to OH levels**

As described in Section 2.2, we perform two additional simulations for low and high OH levels (i.e., S1 and S2) for 1980-2017 to investigate the sensitivity of methane predictions to different OH levels. For both OH cases, the interannual variations in OH levels are the same as in S0 because the simulations are driven by the same meteorology. Figures 13(a) and (b) show global tropospheric OH concentrations and methane OH loss and methane tropospheric lifetime for the three cases (i.e., S0, S1, and S2) in which wetland emissions are optimized (Wopt; Aopt shows a very similar global OH trend as Wopt). Compared to S0, scaling LNOx production in the model by a factor of 0.5 leads to a reduction in simulated global mean OH levels (i.e., by an annual mean of 6.4 %) in S1 and by a factor of 2 leads to an increase in simulated global mean OH levels (i.e., by an annual mean of 9.1%) in S2 over 1980-2017. The global mean OH levels increase from 1980 to 2008 (by an annual mean of 3.6%, with respect to 1980 level) with a linear increasing rate of 4.1×103 molecule cm-3 yr-1, a decrease from 2008 to 2015 (by an annual mean of 2.3%, with respect to 2008 level) with a linear decreasing rate of 7.1×103 molecule cm-3 yr-1, and an increase from 2015 to 2017 (by an annual mean of 4.6%, with respect to 2015 level) with a linear increasing rate of 3.2×104 molecule cm-3 yr-1. However, compared to the 1998-2007, OH levels during 2008-2015 and 2015-2017 are still higher by 2.5% and 1.3%, respectively. Changes in OH levels depend on a number of factors (e.g., temperature, water vapor, O3, NO*x*, CO, and VOCs). Therefore, it is closely related to specific model chemistry and forcing data used for the model. Since emission optimization is also based on methane sinks, the total optimized emissions in S1 are lower than those in S0 by about 4.1% (with an annual mean of -23.7 Tg yr-1), and the total optimized emissions in S2 are higher than those in S0 by about 5.8% (with an annual mean of 33.4 Tg yr-1). This indicates 1% change in OH levels could lead to about 4 Tg yr-1 difference in the optimized emissions. Increasing methane loss due to OH is simulated for 1980 to 2007 in three cases due to increases in OH and methane concentrations (except over the stabilization period when methane was not increasing but OH was increasing). During 2007-2013, the simulated decrease in OH levels combined with increasing methane concentrations lead to relative stabilization in methane OH loss in three cases. The large interannual variability in OH levels during 2013-2017 dominates the interannual variability in methane OH loss despite the continued increases in methane.

All three simulations show a similar trend for tropospheric methane lifetime, with a linear decreasing trend from 1980 to 2007 (i.e., -0.04 year yr-1 in S0, -0.05 year yr-1 in S1, and -0.03 year yr-1 in S2), a clear increasing trend during 2011-2015 (0.08 year yr-1 in all three simulations), and a decreasing trend during 2015-2017(i.e., -0.2 year yr-1 in all three simulations). The mean (for 1980-2017) tropospheric methane lifetime due to OH loss is 9.9±0.4 years in S0Wopt, which is about 0.5 year lower than S1Wopt (i.e., 10.4±0.5 years), and about 0.7 year higher than S2Wopt (i.e., 9.2 ±0.3 years) due to different OH levels and therefore methane sinks, but with similar methane burdens. This indicates 1% change in OH levels could lead to about 0.08 year difference in the tropospheric methane lifetime. The mean tropospheric methane lifetimes simulated by three simulations are within the uncertainty range of observation-derived estimates for the 2000s (Prather et al., 2012) and model estimates (Voulgarakis et al., 2013; Naik et al., 2013). All simulations show an increase in methane lifetime during 2011-2015, which could be a signal of the methane feedback on its lifetime (Holmes, 2018) in the model. Continued increases in methane emissions (Figure 7) during this time period along with decreases in tropospheric OH concentrations (Figure 13) prolong methane lifetime and therefore amplify methane’s response to emission changes. If methane emissions continue to increase, we can expect stronger increases in atmospheric methane due to the amplifying effect of methane-OH feedback, which is also shown in the significant increases in methane growth rates during 2014 and 2015.

**4 Conclusions**

In this work, we thoroughly evaluate the methane budget simulated by the GFDL-AM4 and apply the model to quantify changes in global methane budget. We simulate the DMF of methane and related tracers for 1980 to 2017 by driving the model with gridded emissions inventory compiled from various sources. In order to match the long-term record of surface methane measurements, we optimize global total methane emissions using a simple mass-balance approach. Our optimized global total methane emissions are within the range of estimates by previous studies (both bottom-up and top-down). The GFDL-AM4 simulations with emissions following two different optimizations (anthropogenic and wetlands) are generally able to reproduce observed methane growth rates for the different time periods (e.g., 9.4 ppb yr-1 during 1984-1998, 1.1 ppb yr-1 during 1999-2006, and 6.1 ppb yr-1 during 2007-2017). The simulations are also able to capture the spatial and vertical distribution of methane as retrieved by satellites and measured from aircraft. Our model evaluation show that both simulations with emission optimization on anthropogenic sectors and wetland sector are able to reproduce observed global methane trend and variability despite the different contributions from anthropogenic and wetland emissions. This therefore suggests that the accurate estimates of global total emissions and in the interannual/seasonal variabilities of the emissions are critical in predicting the global methane trend and its variability despite uncertainties in the estimates of individual sources.

We then explore and attribute the methane trends and variability over 1980 to 2017 to sources and sinks. The simulation with emission optimization on anthropogenic sectors shows anthropogenic emissions are the major contributors to the rapid methane growth during 1980s and 1990s whereas the simulation with emission optimization on wetland sector shows wetlands are the major contributors during these periods. However, both simulations suggest increases in methane sources (mainly from energy and waste sectors) balanced by the increases in methane sinks (mainly due to increases in OH levels) leading to methane stabilization during 1999-2006, and energy sector is the major contributor to the methane renewed growth after 2006.

Two additional sensitivity simulations are conducted to further investigate the contributions of wetlands to the methane renewed growth during 2007-2014. The simulation with repeating 2006 emissions for all the sectors except wetland suggests increases in wetland emissions alone are not able to explain the renewed methane growth because sinks are equally important for determining the trend under constant anthropogenic emissions condition. The simulation with combined optimizations (i.e., 1980-2005 optimized anthropogenic emissions and 2006-2014 optimized wetland emissions) suggest the need for a significant increase in wetland emissions along with decreases in anthropogenic emissions starting in 2006 compared to the stabilization period (i.e., 1999-2006) for wetland emissions to drive renewed growth in methane, which is a less likely scenario.

Two additional sensitivity simulations with low and high OH levels (by scaling LNOx production in the model by a factor of 0.5 and 2) are also conducted to further investigate methane OH loss and tropospheric lifetime. In general, OH level trends dominate methane OH loss trends during different methane growth periods except 2007-2013, when methane OH loss shows little change due to the decrease in OH levels combined with increases methane concentrations. The results also indicate 1% change in OH levels could lead to about 4 Tg yr-1 difference in the optimized emissions and 0.08 year difference in the estimated tropospheric methane lifetime. The increasing methane lifetime during 2011-2015 in all the OH sensitivity simulations indicate possible methane feedback on its lifetime in the model. Continued increases in methane emissions along with decreases in tropospheric OH concentrations prolong methane lifetime and therefore amplify methane’s response to emission changes.

Essentially, the global atmospheric methane trend is driven by the competition between its emissions and sinks. When emissions dominate sinks, it leads to an increasing trend, and when sinks dominate emissions, it leads to a decreasing trend. Our model results suggest that the methane stabilization during 1999-2006 is mainly due to increasing emissions balanced by increasing sinks whereas the methane renewed growth during 2007-2013 is mainly due to increasing sources combined with little change in sinks despite small decreases in OH levels. The significant increases in methane growth during 2014-2015 are mainly due to increasing sources combined with decreasing sinks. Most of the model simulations conducted here suggest that increases in energy sources drive the renewed methane growth, in agreement with previous studies (e.g,, Rice et al., 2016; Hausmann et al., 2016; Worden et al., 2017), but in disagreement with other studies that consider emissions from microbial sources as the major contributor (e.g., Nisbet et al., 2016; Schaefer et al., 2016). However, optimization of emissions from anthropogenic sources depends on the “shares” of individual anthropogenic sectors in the initial emission inventories. Uncertainties in these “shares” could lead to uncertainties in the emission adjustment for each anthropogenic sector. Recent studies using isotopic composition suggest that renewed growth in methane is more likely due to the increases in biogenic sources (e.g., Schaefer et al., 2016) as the ratio is shifting to more negative values since 2007. However, it also implies increases in more isotopically lighter fossil fuel emissions, or decreases in isotopically heavy methane source (e.g., biomass burning), or increases in both microbial and fossil fuel emissions but with increases in microbial emissions stronger than those from fossil fuel sources (Nisbet et al., 2019). It is quite possible that the increases in agriculture and waste sectors may drive the renewed methane growth if not energy sector. In that case, it is possible that the emission growth of agriculture and waste sectors could be underestimated in the optimized emissions while the emission growth of energy sector could be overestimated.

The optimized emission totals estimated in this work represent temporal and spatial distribution of methane total sources reasonably well. However, the emission adjustments are either applied to anthropogenic sectors only or wetland sector only. Uncertainties therefore exist on the distribution of the emission adjustments into individual sectors. Without accurate estimates of emissions from individual sources, it would be difficult to attribute the methane trend and variability into specific source. The application of methane isotopes and additional observational constraints (e.g., ethane and δ13CH4) could potentially help better partition the emission adjustments to different sectors. In addition, the spatial distribution of optimized emissions depends on the spatial information from the initial emission inventories. Uncertainties in the spatial distribution from the initial emission inventories could pass to the optimized emissions. Based on the model evaluation, there is possible overestimation of tropical emissions. A process-based emission model (e.g., wetland emissions) could be coupled with AM4 to better represent the spatial and temporal patterns of the emissions.

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