



Importance of emissions-driven simulation of methane for representation of key atmospheric oxidants

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Abstract: Methane (CH₄) is a major greenhouse gas influencing both climate and atmospheric chemistry through its interactions with hydroxyl radicals (OH) and ozone (O₃). Using the FRSGC/UCI chemical transport model, we compare a new emission-driven simulation with configurations where CH₄ mixing ratios are prescribed at the surface or throughout the troposphere. The emission-driven simulation better reproduces the observed spatiotemporal characteristics of CH₄ and captures stronger positive trends over major source regions. With a global CH₄ source of 602 Tg·yr⁻¹ we find sinks of 515 Tg·yr⁻¹ from tropospheric chemical loss and ~36 Tg·yr⁻¹ each from soil uptake and stratospheric loss. The impact of CH₄ treatment on global oxidants is modest, with tropospheric O₃ and OH burdens 0.6% and 0.05% higher in the emission-driven simulation, respectively. The simulated CH₄–OH feedback is insensitive to the choice of CH₄ configuration, with conventional feedback factors ranging from 1.403 to 1.409 across the three configurations. However, there is a pronounced influence on the longer-term response to emission perturbations. A 20% reduction in surface NO emissions produces immediate decreases in O₃ and OH, but the emission-driven simulation additionally captures the persistent response in CH₄ burden, which peaks at 21.5 Tg in the following year and decays with a perturbation lifetime of 12.5 years. The elevated CH₄ enhances O₃, partially offsetting its initial decrease. These results demonstrate that an emission-driven treatment provides a more complete framework for quantifying long-term CH₄–OH–O₃ responses to emission changes and assessing their impacts on atmospheric chemistry and climate.

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Keywords: Methane; Atmospheric chemistry model; Emissions; Boundary condition; Feedback

1 Introduction

Methane (CH₄) is the second most important anthropogenic greenhouse gas after carbon dioxide, with a global warming potential approximately 28–34 times greater on a 100-year time horizon (Myhre et al., 2013). Its atmospheric concentration has more than doubled since pre-industrial times, making it a major driver of climate change (Dlugokencky et al., 2011; Lan et al., 2025). Beyond its direct radiative forcing, CH₄ also influences atmospheric chemistry by affecting tropospheric ozone (O₃) and stratospheric water vapor, contributing additional indirect climate effects (Holmes et al., 2013; Turner et al., 2016; Stevenson et al., 2020). CH₄ has both natural and anthropogenic sources. Natural emissions are dominated by wetlands, which account for 24–29% of the global total source of global



emissions (Zhang et al., 2023; Saunio et al., 2025). Recent satellite-based and inversion analyses have highlighted that tropical wetlands—particularly in the Congo Basin, Amazon, and Southeast Asia—are releasing substantially more CH₄ than previously estimated, playing a key role in the renewed growth of atmospheric CH₄ since 2007 (Bloom et al., 2017; Wilson et al., 2021; Murguía-Flores et al., 2023; Lin et al., 2024). Anthropogenic contributions stem primarily from agriculture, fossil fuel production, and waste management (Jackson et al., 2020; Nisbet et al., 2023; Saunio et al., 2025). The spatial and temporal variability of these emissions, combined with large uncertainties in sinks such as soil uptake and oxidation by hydroxyl radicals (OH), complicates accurate CH₄ budget assessments (Holmes et al., 2013; Nisbet et al., 2019).

Regional CH₄ emissions are uncertain due to sparse observational constraints. Ground-based monitoring networks remain limited in some regions, especially the tropics, while satellite products, though improving, still face retrieval and coverage limitations (Jacob et al., 2022; Qu et al., 2021). Despite these challenges, progress in satellite remote sensing and atmospheric inversions has enhanced our ability to quantify emissions and identify regional hotspots (Bloom et al., 2017). However, discrepancies between top-down and bottom-up approaches remain a central uncertainty in quantifying the CH₄ budget (Saunio et al., 2020, 2025; Jackson et al., 2020).

In chemical transport model (CTM) and earth system model (ESM) studies, CH₄ is most commonly included by prescribing surface mixing ratios. This ensures that concentrations match observations and do not drift under the effects of uncertainty in emissions or biases in chemical removal associated with imperfect representation of OH. Ten of the fourteen global models that contributed to the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) and all models that performed hindcast runs for Phase 6 of the Coupled Model Intercomparison Project (CMIP6) adopted this approach (Voulgarakis et al., 2013; Griffiths et al., 2021). Beyond CH₄, the approach is widely adopted for a range of other long-lived gases and the observational constraint it provides is a major advantage. However, CH₄ boundary conditions are typically provided in discrete latitude bins, and thus zonal variability and emission hot spots are not represented.

Emission-driven simulations offer a more process-based representation of CH₄ sources and enable the geographical attribution of atmospheric CH₄ and its trends to different source regions and emission sectors (Szopa et al., 2013; Stanevich et al., 2020; Folberth et al., 2022). While emission-driven simulations have been shown to be viable (e.g. Patra et al., 2011), a major challenge with this approach is that simulated CH₄ concentrations drift from reality due to biases in the emissions and weaknesses in model representation of tropospheric OH, the primary CH₄ sink. To address this some studies have implemented online ‘flux adjustment’ schemes by altering the magnitude of CH₄ emissions to ensure that mixing ratios remain close to those observed (e.g., Folberth et al. 2022).

Another challenge in driving atmospheric models with CH₄ emissions is the feedback of CH₄ on OH and O₃. CH₄ oxidation consumes OH, which alters the oxidative capacity of the troposphere (Stevenson et al., 2020; Zhao et al., 2020; Zhao et al., 2025). As OH is the primary sink for CH₄, this process constitutes a chemical feedback that influences CH₄ atmospheric lifetime and contributes to nonlinearities in its projected concentrations (Stecher et al., 2021). Additionally, CH₄ oxidation leads to formation of formaldehyde and carbon monoxide (CO), which can enhance tropospheric O₃ formation, particularly under polluted conditions (Voulgarakis et al., 2013; Turner et al., 2016). Holmes (2018) compared CH₄ lifetime and feedback under three approaches to representing CH₄ and found that differences in feedback factors were very small, although an emission-driven configuration exhibits the strongest feedback. Emissions of hydrocarbons and CO were shown to weaken the CH₄ feedback, whereas nitrogen oxide (NO_x) emissions have a more complex effect, reducing CH₄



85 feedback over source regions but enhancing it elsewhere. In contrast, Voulgarakis et al. (2013) reported that declining anthropogenic NO_x emissions contribute to a projected strengthening of the CH₄ feedback through the twenty-first century under the RCP8.5 scenario.

Global atmospheric chemistry model simulations consistently show that decreases in NO_x emissions reduce OH concentrations and prolong the global methane lifetime (Derwent et al., 2001; Wild et al., 2001; Stevenson et al., 2004; Weber et al., 2020). Although CH₄ has an atmospheric lifetime of roughly 10 years, model studies demonstrate that it responds rapidly to sudden, short-lived NO_x emission pulses within a few months (Wild et al., 2001; Stevenson et al., 2004). Stevenson et al. (2022) further demonstrated that the abrupt emission changes during the COVID-19 lockdowns rapidly affected the global CH₄ growth rate, with NO_x emission reductions contributing to an increase of 4.8±1.0 ppb·yr⁻¹. While the overall methane feedback is reasonably well constrained, the coupled CH₄–O₃–OH response to emission perturbations remains uncertain and requires further quantitative investigation.

Given the uncertainties and limitations associated with CH₄ sources, sinks, and its atmospheric behaviour, it is essential to understand how different approaches to representing CH₄ in models influences its simulated impacts. This study aims to quantify the effects of three commonly used frameworks—fixed field, fixed surface and emissions-driven approaches—on the spatial and temporal distribution, atmospheric lifetime, and chemical feedback of CH₄, particularly in relation to key oxidants such as OH and O₃ and its response to NO_x emissions. Using an updated chemical transport model, we conduct a series of simulations to compare these approaches. Section 2 describes the model setup, simulation design, and observational datasets used for evaluation. Model evaluation, differences in CH₄ budget and trend, impact on tropospheric oxidants, CH₄ chemical feedback, and response to surface NO_x emissions are presented in Section 3, followed by conclusions in Section 4.

2 Model and methods

In this section, we provide a brief overview of the CTM in its standard configuration with prescribed 3-D fields of CH₄ and the modifications introduced in this study to implement additional configurations based on fixed surface concentrations and on CH₄ emissions.

2.1 Chemical Transport Model

The global atmospheric model used here is the Frontier Research System for Global Change version of the University of California, Irvine (FRSGC/UCI) CTM. The model is described in Wild and Prather (2000) and has been widely used to study the tropospheric chemistry and transport of trace gases including O₃ and OH (e.g. Wild et al., 2020). Results from the model have been included in a range of model intercomparison initiatives, such as the Hemispheric Transport of Air Pollution (HTAP) projects (Fiore et al, 2009). The model was recently updated and extensively evaluated by Hou et al. (2026). The CTM uses offline meteorological data generated with the European Centre for Medium-Range Weather Forecasts (ECMWF) Integrated Forecast System (IFS) version Cy38r1.

2.2 Chemistry and Emissions in the Default Configuration

The default configuration of the model used here is the same as that described and evaluated by Hou et al. (2026). Briefly, it includes explicit treatment of HO_x/O_x/NO_x chemistry, CH₄ and isoprene oxidation, and a lumped approach for other non-methane hydrocarbons (NMHCs) based on representative species butane, propene and xylene (Wild, 2007). Alongside gas-phase chemistry, the model now includes a detailed treatment of heterogenous chemistry occurring on tropospheric aerosols, cloud liquid water and cloud ice (Hou et al., 2026).



Monthly emissions of CO, NO_x and NMHCs are prescribed largely from inventories produced by the Copernicus Atmosphere Monitoring Service (CAMS). Anthropogenic emissions are from version 6.2 of the CAMS-GLOB-ANT inventory (Soulie et al., 2024), aircraft emissions from version 2.1 of CAMS-GLOB-AIR (Granier et al., 2019), biogenic emissions from version 3.1 of CAMS-GLOB-BIO (Sindelarova et al., 2022) and soil emissions from version 2.4 of CAMS-GLOB-SOI (Simpson and Darras, 2021). Fire emissions are taken from the Global Fire Emissions Database, Version 4.1 (GFED4s) inventory (van der Werf et al., 2017). Each of the emissions above vary annually over our full simulation period. Oceanic emissions of CO and selected NMHCs are taken from the POET emission inventory and are assumed to be climatological (Granier et al., 2005). Lightning NO_x emissions are included in the model and vary annually, providing a source of about 5 Tg[N]·yr⁻¹.

2.3 Modifications to Represent CH₄ Sources and Sinks

In the standard model simulations, an annual and latitudinally varying mixing ratio of CH₄ is prescribed using the historical forcing data prepared for CMIP6 until 2014 (Meinshausen et al., 2017). Post 2014, to match the observed record, the fields are scaled by a globally uniform factor derived from CH₄ measurements of the National Oceanic and Atmospheric Administration (NOAA) global monitoring network. In this standard configuration of the model (STD_Fix), the CH₄ mixing ratios are prescribed uniformly through the depth of the troposphere. For an alternate configuration (STD), we prescribe CH₄ mixing ratios at the surface only and allow model transport processes to define concentrations above the surface.

For an emission-driven configuration (CH4E), we have replaced the prescribed CH₄ mole fractions with explicit monthly emissions. Six CH₄ source types are included: anthropogenic, wetland, geological, termite, fire, and biogenic emissions. The annual global emission totals are shown in **Figure 1**, and the spatial distribution is shown in **Figure S1**. For the anthropogenic source, we adopt the CH₄ emissions inventory produced for CMIP7 (Hoesly et al., 2025). Wetland emissions are from the high emissions scenario in the extended ensemble subset of Bloom et al. (2017). Fire emissions are from the GFED4s inventory (van der Werf et al., 2017). Biogenic emissions are from CAMS-GLOB-BIO (v3.1) inventory (Sindelarova et al., 2022). Emissions from termites are taken from the CAMS-GLOB-TERM (v1.1) inventory and do not vary annually but are assumed representative of the period 2000 – 2017 (Granier et al., 2019). Estimated geological emissions for the year 2022 are taken from Martinez et al. (2024) and Saunio et al., (2025), and are fixed in time due to limited information on annual variability.

The dominant chemical sink of CH₄ in the atmosphere is reaction with OH, which is responsible for 80-90% of the total chemical CH₄ loss (Kirschke et al., 2013; Saunio et al., 2025; Folberth et al., 2022). The temperature-dependent rate coefficient for this reaction is taken as $2.45 \times 10^{-12} \exp(-1775/T)$ cm³·molecules⁻¹·s⁻¹ (Burkholder et al., 2020). Chlorine radicals provide an additional CH₄ sink, ~0.8% of global CH₄ oxidation (Wang et al., 2021), but this is relatively small and is not included in the model at present. The chemical removal of CH₄ in the stratosphere is parameterized using a first-order loss rate above the tropopause, and the CH₄ global lifetime to stratospheric loss is approximately 140 years. This estimate is consistent with previous estimates of 120 years in Prather et al. (2012) and 195 (153–270) years in Brown et al. (2013).

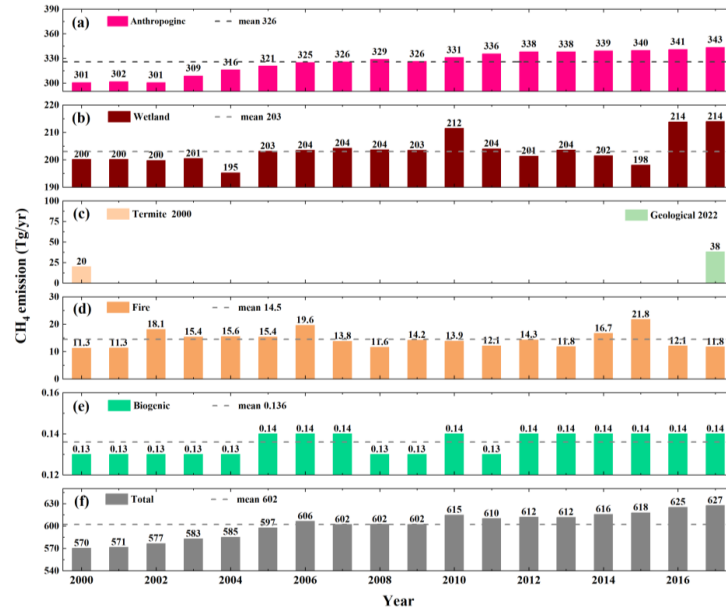


Figure 1. Annual CH₄ emissions (Tg/yr) implemented in the CH₄E configuration of the model (2000-2017). The bottom row (f) shows the global total emissions while rows above show the individual contributions from (a) anthropogenic, (b) wetland, (c) geological and termite, (d) fire, and (e) biogenic sources. In each panel, the dashed horizontal line indicates the mean emission rate over 2000–2017. Geological and termite emissions are time-invariant.

Table 1. Typical values of CH₄ dry deposition velocities

Ecosystem type	V_d ($\times 10^{-6} \text{ m s}^{-1}$)
Ice and water	0
Tropical forest	10.6
Boreal forest	8.5
Grassland	7.4
Tundra	4.8
Desert	3.5
Cultivation	3.9

For the emissions-driven configuration, we implemented surface soil uptake based on the CH₄ dry deposition velocity. Typical values of CH₄ dry deposition velocities over different land types are shown in **Table 1** and are calculated from Equation (1) based on the mean uptake rates of CH₄ from Dutaur and Verchot (2007).

$$V_{di} = \frac{F_i}{F} \times V_{d0} \times \frac{1}{r} \quad (1)$$



V_{di} is the CH₄ dry deposition velocity to land type i ; F_i is the mean uptake rate over land type i in Dutaur and Verchot (2007); F is the total mean uptake in Dutaur and Verchot (2007); V_{d0} is the global mean dry deposition velocity, taken here as $3.2 \times 10^{-6} \text{ m}\cdot\text{s}^{-1}$. r is the ratio of land area to the global surface area. Following the approximate global land fraction reported in Zhao et al. (2017), r is taken as 0.30 in this study.

2.4 Experimental approach

We performed three model experiments, STD_Fix, STD and CH4E, the main features of which are summarized in **Table 2**. All runs were performed at T42 horizontal resolution ($2.8^\circ \times 2.8^\circ$) and with 57 vertical levels extending from the surface to 0.1 hPa. The experiments used the same meteorological conditions and the same emissions for species other than CH₄. CH₄ mixing ratios were prescribed everywhere in STD_Fix, only in the lowest layers at the surface in STD, and CH₄ emissions were used in CH4E. All experiments were run for the period 2000 to 2017. Preceding this, a spin-up of 2.5 years was used for STD_fix and STD experiments, and a longer spin-up of 7.5 years was used to balance the impact of CH₄ emissions (CH4E) on other species at the start of the simulation.

Table 2. Experimental settings

	Run STD_Fix	Run STD	Run CH4E
CH ₄ boundary conditions	Prescribed mole fractions everywhere (Fixed field)	Prescribed mole fractions at the surface (Fixed surface)	6 sources, total ~602 Tg/yr (specified emissions)
Year	2.5-yr Spin-up; 2000-2017	2.5-yr Spin-up; 2000-2017	7.5-yr Spin-up; 2000-2017
CH ₄ Deposition	Updated deposition scheme in section 2.3		
Stratospheric loss	First-order loss with a rate constant of 0.05 yr^{-1}		
Meteorology	ECMWF Integrated Forecasting System (IFS) model versions Cy38r1		
Emissions	CAMS-GLOB+GFED4S fire		
Resolution	T42L57		

2.5 Observation data

To evaluate model performance, we use monthly surface CH₄ measurements and vertical CH₄ profiles obtained by the National Oceanic and Atmospheric Administration (NOAA) Global Monitoring Laboratory (GML). A total of 67 surface observation sites with continuous CH₄ records spanning the study period (2000-2017) were selected. The site information is provided in **Table S1**, and further details are available in Lan et al. (2025).

Vertical CH₄ profile data were obtained through NOAA GML's Aircraft Program, which supports a range of measurement and monitoring activities (McKain et al., 2026). These include routine airborne sampling at fixed sites across the USA under the NOAA GML's Global Greenhouse Gas Reference Network (GGGRN), targeted intensive campaigns, and the National Observations of Greenhouse gases Aircraft Profiles (NOGAP) program. The NOGAP program conducts near-continuous atmospheric sampling around the continental USA with the goal of addressing spatial sampling gaps. In total, vertical profile data from 33 observation sites covering the study period were used. Site-specific information is summarized in **Table S2**.

3 Results

3.1 Model evaluation

Figure 2a compares modeled and observed surface CH₄ mixing ratios at 67 NOAA sites, averaged



over the period 2000–2017, as a function of latitude. Both prescribed and emission-driven configurations generally capture the latitudinal gradient of CH₄ well, with higher concentrations in the northern hemisphere than the southern hemisphere, especially between 30°N and 60°N, where emissions are more intense (**Figure S2**). Model biases for the prescribed and emission-driven runs are shown in **Figures 2b** and **2c**, respectively. Surface CH₄ concentrations in prescribed concentration runs (STD and STD_Fix) are the same and thus the simulations are not shown separately here. The STD prescribed concentration simulation has a mean bias of -5.7 ± 27.9 ppbv (mean $\pm$ 1 standard deviation across the 67 sites), with particularly large negative biases at some sites in the northern hemisphere mid-latitudes (30°N–60°N). This underestimation may arise from the assumption of annual and zonal mean concentrations along latitude. The uncertainty may also be associated with extending the prescribed CH₄ concentration data to 2015–2017. The emission-driven CH₄E simulation has a positive mean bias of 14.2 ± 19.6 ppbv, but a smaller spatial spread in site-level biases, indicating that explicitly resolving CH₄ emissions reduces some of the large regional discrepancies found in the prescribed concentration simulation. While this simulation overestimates CH₄ in some regions, especially at mid- to high- latitudes, it better reproduces the observed interhemispheric gradient. For example, the observed interhemispheric gradient from the sites in **Figure 2a** is 102.5 ppbv, compared with 94.7 ppbv in the STD run and 100.4 ppbv in the CH₄E run.

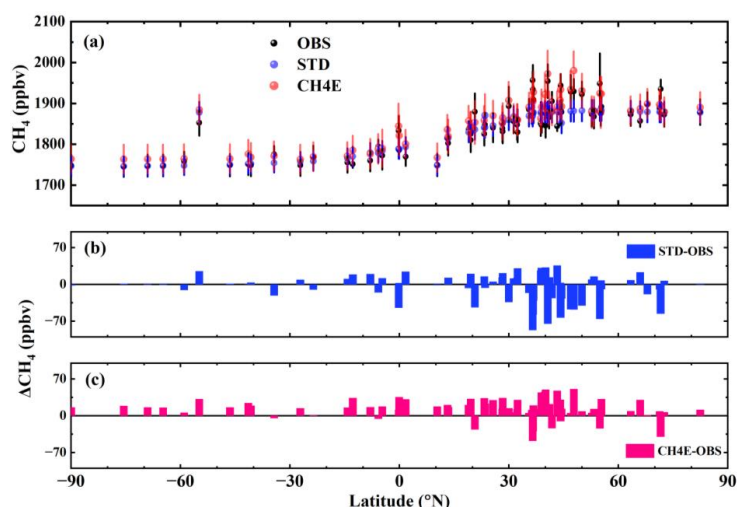


Figure 2. (a) Latitudinal distribution of observed and modeled surface CH₄ (ppbv) averaged over the period 2000–2017. The observations are from the NOAA GML network (OBS, 67 sites). Model output is shown for STD (fixed surface) and CH₄E (emission-driven) runs. (b) and (c) Corresponding model minus observation difference in CH₄ (ppbv). Positive values indicate overestimation by the model.

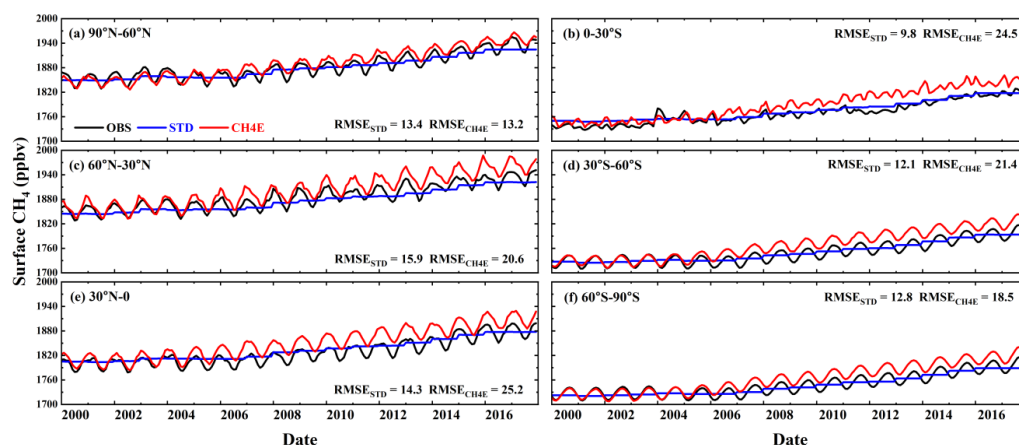


Figure 3. Time series of monthly mean surface CH₄ (ppbv) from emission-driven (CH4E) and prescribed (STD) simulations against observations in different latitude bands. The root mean square error (RMSE) between the model and observations (ppbv) are labelled in each panel.

Figure 3 shows the time series of modelled surface CH₄ mixing ratios from 2000 to 2017, averaged within 30° latitude bands, against observations. The root mean square error (RMSE) for each model run is also shown in each panel. In general, both model runs closely follow the observed CH₄ trends during the early years (2000–2004) across all latitude bands. From around 2005 onward, the emission-driven CH4E simulation begins to show a small systematic positive bias relative to the observations, particularly in the tropics and southern hemisphere, suggesting an overestimation of CH₄ emissions or underestimation of CH₄ sinks in this run. One potential explanation for this discrepancy is the flattening of the OH trend in the model (Hou et al., 2026), which contrasts with the increasing trends observed in other studies. Overall, averaged over our study period the emission-driven simulation yields lower RMSE than the prescribed concentration simulation in the 90°N–60°N band, but higher RMSE in most other latitude bands, particularly in the tropics (30°N–0° and 0°–30°S). However, the magnitude of the biases in the emission-driven simulation are sufficiently small that we do not require an emission flux adjustment of the type described by Folberth et al. (2022).

The emission-driven simulation captures the amplitude and phase of the observed CH₄ seasonal cycle extremely well in all latitude bands (**Figure S3**). The prescribed concentration simulation shows no seasonal variation in the anomalies because annual mean surface CH₄ mixing ratios are prescribed without a seasonal cycle in this configuration.

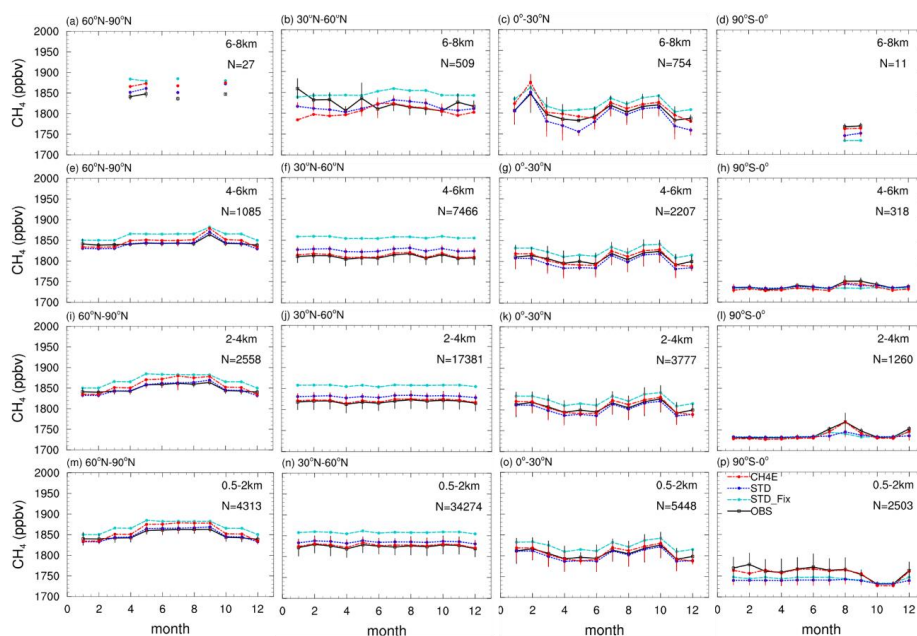


Figure 4. Multi-year averaged monthly variations in simulated and observed CH_4 mixing ratio (ppbv) from 2000 to 2017 for different latitude bands and four altitude ranges: 0.5–2 km, 2–4 km, 4–6 km, and 6–8 km (bottom to top). Observational data are from NOAA's aircraft measurement program using discrete flask-air samples collected at multiple altitudes. N indicates the number of samples used in each dataset.

To further evaluate the vertical and seasonal variations in CH_4 , **Figure 4** compares the simulated and observed monthly CH_4 mixing ratios over 2000–2017 across four latitude bands and altitude ranges. As CH_4 in the fully prescribed simulation is fixed to an annual-mean distribution, the apparent monthly variations in this simulation arise from differences in the spatial and temporal sampling of the observations rather than from simulated seasonal variability. The prescribed-surface and emission-driven simulations generally reproduce the observed variations across the different latitude and altitude ranges, including the large-scale latitudinal gradient, with higher CH_4 mixing ratios in the northern hemisphere than in the southern hemisphere, and the latitude-dependent seasonal variations. The two simulations show similar vertical and seasonal patterns, with the emission-driven simulation generally showing closer agreement with the observed CH_4 mixing ratios. These results further indicate that explicitly resolving CH_4 emissions and sinks can reproduce the observed large-scale vertical, latitudinal, and seasonal characteristics reasonably well.

3.2 CH_4 global budget

The global CH_4 budget averaged over the periods 2000–2009 and 2010–2017 from the three model simulations and from recent literature studies over similar periods (Saunois et al., 2025; Folberth et al., 2022) are compared in **Table 3**. All budget terms diagnosed from the emission-driven CH_4E run are within the range of estimates from bottom-up studies as reported by Saunois et al. (2025). The CH_4E simulations show a higher surface uptake than in the standard prescribed concentration run (35.7 vs. 34.2 Tg/yr), reflecting the zonal asymmetry associated with higher CH_4 over continental regions. The stratospheric loss term is similar for the two runs (35.7 Tg/yr in STD, 35.6 Tg/yr in



CH₄E), both within the range of estimates from other recent studies.

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Table 3. CH₄ budgets (Tg/yr) and lifetime (years) from 2000 to 2017

	Period	Emission	Chem loss (OH)	Surface uptake	Strat Loss	Burden	Total lifetime ⁴	Lifetime due to OH loss
CMIP6 ¹	2000-2009	638 ³	541	30	39	/	8.2	9.3 ⁵
CMIP6 ¹	2010-2019	669 ³	563	31	37	/	8.2	9.2 ⁵
UKESM1- ems ²	2000-2009	591	525	31	23	/	/	8.5±0.1
STD_Fix	2000-2009	/	504	33.8	/	5042	/	9.95
STD_Fix	2010-2017	/	519	34.6	/	5163	/	9.90
STD	2000-2009	/	503	33.8	35.7	4946	8.64	9.78
STD	2010-2017	/	518	34.6	35.6	5049	8.58	9.70
CH ₄ E	2000-2009	590	506	35.1	35.0	4955	8.60	9.74
CH ₄ E	2010-2017	617	524	36.2	36.1	5127	8.60	9.74

¹mean values, Saunio et al. (2025)

²Folberth et al. (2022)

³bottom-up, Saunio et al. (2025)

⁴defined as total burden divided by total loss which includes tropospheric chemical loss, stratospheric removal, and surface uptake to soils.

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⁵tropospheric lifetime defined as total burden divided by tropospheric loss.

The CH₄ total lifetime and lifetime due to OH loss are shown in **Table 3**. The total lifetime is calculated as the global CH₄ burden divided by the total sink where the latter includes removal of CH₄ by oxidation in the troposphere, stratospheric removal, and surface uptake to soils. The total CH₄ lifetime from the emission-driven and prescribed concentration runs are similar (~8.6 years), indicating that there is little sensitivity to the approach used to implement CH₄ in the model. The chemical lifetime due to OH loss is also similar (~9.7 years) but is notably shorter than for the STD_Fix run (9.9 years) where uniform mixing ratios are prescribed throughout the depth of the troposphere. This highlights the importance of representing the vertical profile of CH₄ appropriately.

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3.3 Differences in CH₄ spatial and temporal distribution

The impact of incorporating CH₄ emissions in the CH₄E simulation is clearly apparent in the simulated global surface distribution of CH₄, as expected. (**Figure 5**). Enhanced CH₄ concentrations in the emission-driven simulation are observed in regions with significant anthropogenic emissions, including the USA, Europe, India, East Asia and Southeast Asia. Additionally, natural sources, such as wetland emissions, contribute to higher CH₄ concentrations in areas such as Central Africa. Biomass burning also contributes to high CH₄ concentrations over Central Africa. In the Arctic, higher levels of CH₄ are simulated in the CH₄E run due to transport from lower latitudes, particularly from sources over Eurasia (Liu et al., 2025).

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The two prescribed-concentration simulations (STD_Fix and STD) have the same prescribed surface CH₄ distribution, but differ in their vertical distribution: CH₄ is prescribed throughout the atmosphere in STD_Fix, but only at the surface in STD. Consequently, STD develops a vertical CH₄ gradient



through atmospheric transport and chemical loss, similar to that in the emission-driven CH₄E simulation (**Figure 5c, d**). Both STD and CH₄E show a gradual decrease in CH₄ from the surface to the tropopause, with broadly similar vertical gradients, particularly in the northern mid- and high latitudes where CH₄ abundances are highest. CH₄ concentrations are generally higher in CH₄E than in STD, reflecting differences arising from the explicit treatment of surface emissions and atmospheric sinks. Overall, the emission-driven treatment has a relatively small effect on the large-scale vertical gradient of CH₄, which is primarily controlled by atmospheric transport and chemical loss, although regional differences can arise from the spatial distribution of emissions and their interaction with transport.

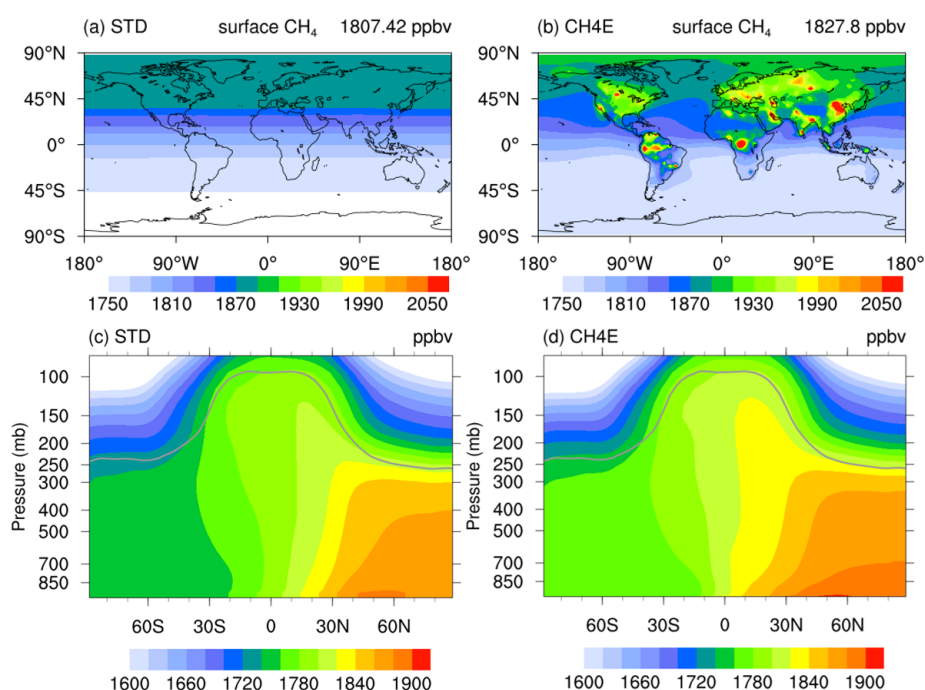


Figure 5. Multi-year averaged annual surface distributions and vertical profiles of CH₄ mixing ratio (ppbv) from 2000 to 2017, derived from (a, c) STD and (b, d) CH₄E simulations. Values in the upper right corner of the surface panels represent the global mean concentrations. The grey line in the vertical panels indicates the tropopause, defined by a 150 ppbv O₃ concentration.

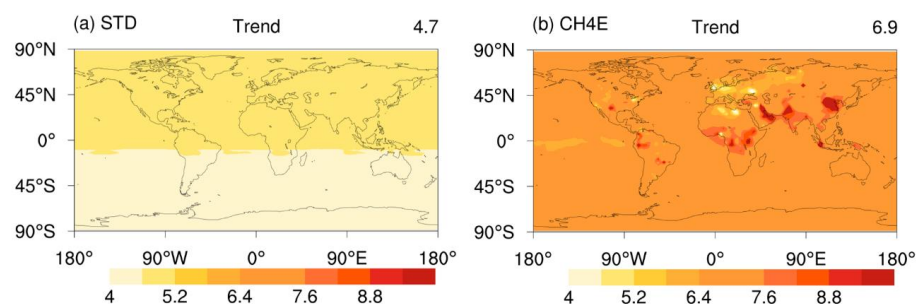


Figure 6. Spatial distribution of annual trends in surface CH₄ during 2000–2017 (ppbv·yr⁻¹) in the (a) STD and (b) CH₄E model runs. Trends were derived using the Theil–Sen estimator. Statistically significant trends at the



330 95% confidence level were assessed using the Mann-Kendall test. The trends are statistically significant across the entire global dataset. The value in the upper right corner of each panel represents the global mean trend in ppbv·yr⁻¹.

In comparing concentration and emission-driven approaches to implementing CH₄, another key issue is if and how long-term trends differ on a regional basis. The growth rate of atmospheric CH₄ in the prescribed concentration simulation is 4.7 ppbv·yr⁻¹ (**Figure 6a**), consistent with the NOAA GML observations, which show a linear increase of 4.58 ± 0.39 ppbv·yr⁻¹ during 2000–2017 (Lan et al., 2026). From **Figure 6**, clear differences in the simulated CH₄ trend are apparent between the prescribed concentration and emission-driven runs, with the latter exhibiting much greater spatial structure. Stronger positive trends are observed in regions such as Africa, South America, India, East Asia and Southeast Asia. For example, CH₄ trends exceed 10 ppbv·yr⁻¹ over northern China, while remaining below 4 ppbv·yr⁻¹ over Europe, indicating a more than twofold difference between these regions. These enhanced trends are likely driven by significant increases in CH₄ emissions in these areas and reflect an improved sensitivity to regional emission sources and their impact on global CH₄ concentrations and trends in the emission-driven run.

3.4 Impact on tropospheric O₃ and OH

The tropospheric O₃ and OH budget from the different runs are summarized in **Table 4**. In the emission-driven CH₄E run, the tropospheric O₃ burden is ~2.0 Tg (0.6%) larger than in the prescribed concentration STD run, and the OH burden is 0.1 Mg (0.05%) larger. Despite the higher O₃ burden, the tropospheric O₃ lifetime remains largely unchanged. The sensitivity of the O₃ and OH global budget terms to the approach to implementing CH₄ is thus very small. In STD_Fix, where concentrations are prescribed uniformly in altitude, the O₃ burden is almost the same as in STD, where concentrations are prescribed at the surface only, but OH is lower due to greater OH reaction with CH₄ due to the higher CH₄ abundance in the upper troposphere.

355 **Table 4.** Tropospheric budgets of O₃ and OH and atmosphere lifetime (days) in the model averaged over the 2000–2017 period.

	O ₃ (Tg·yr ⁻¹)				Trop OH	[OH] _{gm}
	Lifetime	Burden (Tg)	Prod	Loss	Burden (Mg) (Molecules·cm ⁻³)	
STD_Fix	29.1	348.4	5077	4379	208.7	10.86
STD	29.1	348.3	5074	4376	209.0	10.88
CH ₄ E	29.1	350.1	5102	4400	209.1	10.88

In CH₄E, the higher O₃ burden is mainly driven by elevated CH₄ levels, which serve as a key precursor for O₃ formation through oxidation processes. Enhanced CH₄ oxidation promotes O₃ production, as reflected in the production terms shown in **Table 4**. As one of the primary sources of OH is via the photolysis of O₃ (for a chemical budget analysis, see **Table S3**), the higher O₃ concentration under CH₄E contributes to increased OH production. However, CH₄ is also a major sink for OH, and the enhanced CH₄ levels lead to increased OH consumption through oxidation. As a result, the net increase in OH is small due to the competing effects of O₃-driven production and CH₄-driven loss. These results highlight the complex and nonlinear interactions between CH₄, O₃, and OH radicals in the troposphere, and emphasize the importance of considering both source and



sink processes in evaluating atmospheric oxidant budgets.

The surface and zonal mean O_3 distributions from the STD run and the differences between CH4E and STD runs are shown in **Figure 7**. The distributions for the STD_Fix run are similar to those from STD and are not shown here. At the surface, the STD run exhibits the expected inhomogeneous distribution, with annual mean surface O_3 generally higher in the northern hemisphere than in the southern hemisphere, with mixing ratios peaking at approximately 60 ppbv over the Tibetan Plateau (**Figure 7a**). Higher surface O_3 is simulated in the CH4E run over much of the world but the differences are small (globally averaged, +0.2 ppbv; **Figure 7b**). In regions such as East Asia and parts of Europe and North America, larger surface O_3 increases of 0.3 ppbv are apparent due to higher CH_4 concentrations and more efficient O_3 production in VOCs-sensitive regions (Liu et al., 2022). In the vertical, the CH4E run shows a similar structure but exhibits slightly higher O_3 concentrations at higher latitude (**Figure 7c and 7d**), where differences reach 0.4 ppbv in the mid- and upper-troposphere.

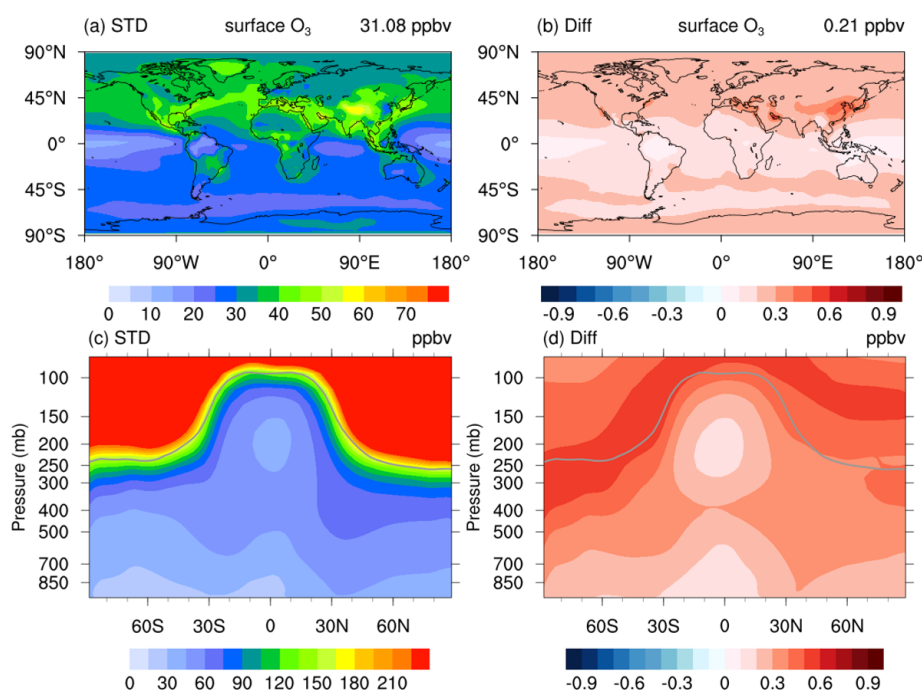


Figure 7. (a, c) Multi-year averaged annual surface O_3 mixing ratio (ppbv) and latitude-pressure O_3 distribution (ppbv) from 2000 to 2017, based on the STD simulation. (b, d) Corresponding differences between the CH4E and STD simulations. The values in the upper right corner of the surface O_3 panels indicate the global mean mixing ratios. The grey line in the vertical distribution panels marks the tropopause, defined by an O_3 mixing ratio of 150 ppbv.

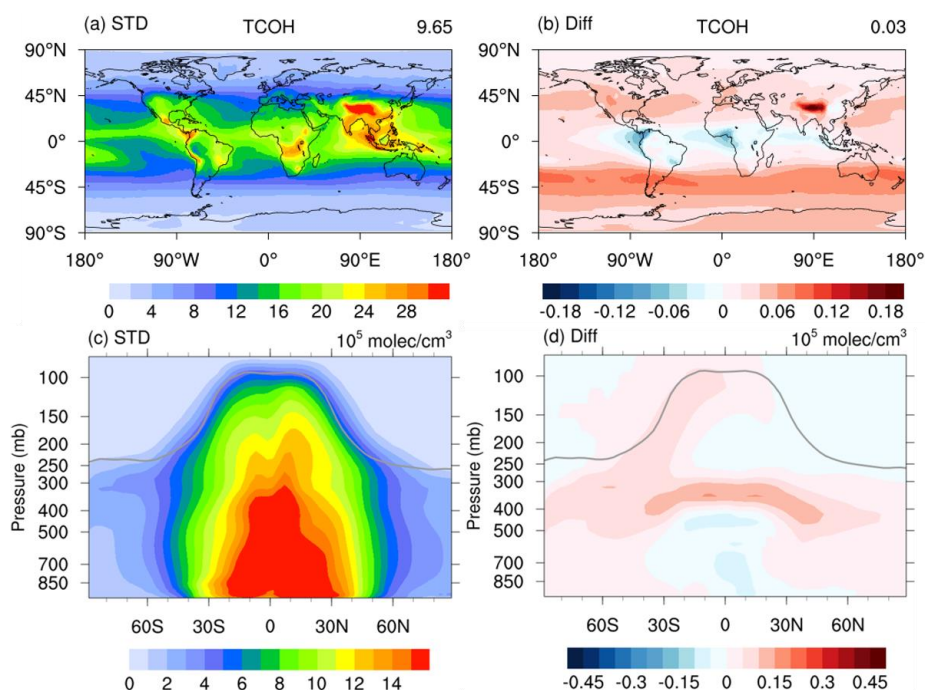


Figure 8. As Figure 7, but for annual tropospheric OH column (TCOH, 10^{12} molecules·cm⁻²) and [OH] vertical distribution (10^5 molecules·cm⁻³).

390 The global distribution of the tropospheric OH column (TCOH) and the zonal mean OH concentrations averaged over 2000–2017 are shown in **Figure 8**. In the STD run, the OH column exhibits a pronounced spatial pattern, with elevated values primarily located in tropical and subtropical regions such as Southeast Asia, central Africa, and northern South America. These regions coincide with strong solar radiation and high humidity, which favor O₃ photolysis and subsequent OH formation. The global mean OH column is 9.65×10^{12} molecules·cm⁻².

395 The differences in tropospheric OH column associated with the emissions-driven CH₄E run are relatively small ($+0.03 \times 10^{12}$ molecules·cm⁻²), but clear regional differences are observed. Small positive anomalies are found in East and Southeast Asia and over the mid-latitude oceans, while negative anomalies appear over tropical regions, such as South America and Africa. These patterns reflect the dual role of CH₄ in enhancing O₃ formation, which promotes OH production via photolysis, but acting as a direct sink of OH through oxidation. The net effect on OH depends on the local balance of these processes.

405 The zonal mean vertical distribution further reveals that OH concentrations peak in the tropical troposphere (surface to 250 mb), consistent with strong photochemical activity in this region. Under the CH₄E run, OH concentrations are slightly higher by up to 0.2×10^5 molecules·cm⁻³ in the middle troposphere, especially in the tropics where CH₄ contributes to O₃ and subsequent OH production. In contrast, at lower altitudes, tropical and northern hemisphere regions show minor OH reductions due to increased consumption by CH₄ oxidation. Overall, these results highlight the subtle but non-negligible impact of CH₄ on tropospheric OH, with altitude- and region-dependent effects governed



410 by the interplay between OH sources and sinks.

3.5 Impact on OH and O₃ trends

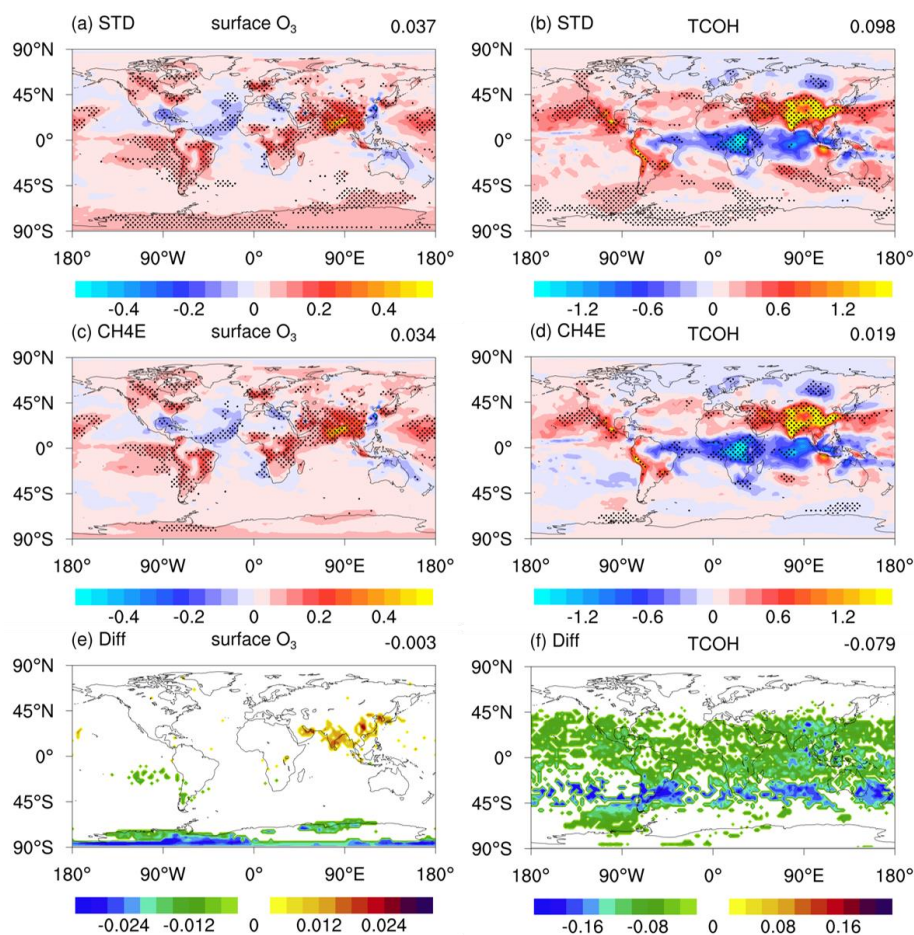


Figure 9. Spatial distribution of annual trends in surface O₃ (ppbv·yr⁻¹, a and c) and tropospheric column OH (10¹¹ molecules·cm⁻²·yr⁻¹, b and d). Panels (e) and (f) show the differences in trends between CH₄E and STD.

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In the prescribed concentration STD simulation, surface O₃ shows small positive trends, especially in Europe, North America, South America, and South Asia (see **Figure 9**). The emission-driven CH₄E run shows similar trends but with slightly larger O₃ increases (~10%) in South and East Asia and smaller O₃ increases at southern high latitudes. The higher O₃ increase in South and East Asia is primarily driven by the high NO_x abundance in these regions, where higher CH₄ concentrations enhance the efficiency of O₃ formation.

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For OH, the STD simulation produces a larger positive trend in the troposphere, particularly in the tropics and northern hemisphere. The CH₄E run shows a slower increase in OH column, particularly in the tropics and Southern Hemisphere mid-latitudes. This is because higher CH₄ in the CH₄E simulation leads to higher CH₄ oxidation, consuming more OH, and reduces the overall OH column growth relative to the STD simulation. In the Southern Hemisphere, TCOH shows a smaller increase

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or even a small decrease that may be associated with the slower O₃ growth in this region.

3.6 Influence of CH₄ treatment on its chemical feedback

Understanding the feedback of CH₄ on its own lifetime is essential for quantifying the response of atmospheric CH₄ to perturbations in its sources and sinks, and plays a critical role in assessing future climate forcing. In this study, we calculate the feedback factor (F) for each of the three approaches we have used to implement CH₄, following the methods described in Holmes (2018). For each approach we perform an additional simulation where the prescribed concentration (for STD and STD_Fix) or emission (for CH₄E) is increased by 20%, and we focus on the responses in the year 2010. We use a 2.5-year spin-up for the prescribed concentration everywhere (STD_Fix) and 14.5-year spin-up for the prescribed concentration at surface (STD) and emissions-driven simulation (CH₄E). We calculate two feedback factors: a conventional feedback factor based on the derived chemical loss to OH and recommended lifetimes for other sinks (stratosphere: 120 years, deposition: 150 years, Cl loss: 200 years) following Prather et al. (2012), and a model feedback factor based on the sinks diagnosed in the model only. The conventional feedback factor permits direct comparison with previous studies, while the model feedback factor characterizes the true behavior of CH₄ in the simulations performed here.

Table 5. CH₄ budget terms and feedback factors simulated with the prescribed and emission-driven CH₄ configurations

	STD_Fix	STD	CH ₄ E
Chemical lifetime (years)	9.93	9.73	9.82
Total lifetime (years)	8.75	8.59	8.62
CH ₄ burden 2010 (Tg)	5013	4999	5144
CH ₄ burden change (Tg)	+1021 (20%)	+1000 (20%)	+1108 (22%)
Conventional feedback factor, F _t	1.4089	1.4094	1.4027
Model feedback factor, F	1.4422	1.4389	1.4394

As shown in **Table 5**, the 20% perturbation in atmospheric CH₄ concentrations in the prescribed concentration simulations resulted in increases in CH₄ burden of 1021 and 1000 Tg. In the emission-driven simulation, a 20% increase in CH₄ emissions produced a slightly larger relative increase in the CH₄ burden, reaching 1108 Tg (22%). Despite these differences, the calculated feedback factors were highly consistent among the three configurations. Using the conventional formulation with model-diagnosed CH₄ chemical loss by OH, together with recommended loss frequencies for Cl oxidation, stratospheric loss, and soil uptake, the feedback factors were 1.4089, 1.4094, and 1.4027 for STD_Fix, STD, and CH₄E, respectively. The differences in the feedback factors among the three CH₄ configurations are therefore small, indicating that the approach to representing CH₄ has only a minor effect on the diagnosed feedback factor. This result is consistent with the findings of Holmes (2018). When the feedback factor was calculated using only the CH₄ loss processes diagnosed directly by the model, excluding Cl oxidation, the corresponding values were 1.4422, 1.4389, and 1.4394, which are slightly higher than those obtained using the conventional formulation. This difference suggests a stronger diagnosed CH₄ feedback when the model-simulated loss processes are used and highlights the sensitivity of the feedback factor to the treatment of individual CH₄ sinks, particularly neglect of Cl oxidation.



3.7 Response to surface NO_x emissions

As well as acting as an important source of key oxidants such as ozone, NO_x emissions have pronounced impacts on the atmospheric CH₄ burden through their influence on tropospheric OH (Wild et al., 2001; Derwent et al., 2001). To explore how the treatment of CH₄ influences assessment of the impacts of NO_x emission changes, we conduct additional simulations with a 20% reduction in surface NO emissions applied in the first year only and follow the subsequent response. We do this for the prescribed surface concentration (STD) and emission-driven (CH₄E) approaches, and perform additional emission-driven runs using repeating year 2000 precursor emissions and meteorology (CH₄E_Fix) to investigate the effects of these factors on the response.

As shown in **Figure 10**, the NO emission reduction induces an immediate and short-lived decrease in O₃ and OH burdens of a similar magnitude in all runs, and these lead to a rapid increase in the CH₄ burden in the emissions-driven run that peaks at 21.5 Tg (CH₄) in 2001, the year after the NO emission reduction. This corresponds to an increase in surface CH₄ abundance of 5.0 ppbv, comparable in magnitude to the 4.8±1.0 ppbv increase seen over the COVID-19 lockdown period in 2020 diagnosed by Stevenson et al. (2022) that was associated with a ~15% NO_x emission reduction. Thereafter, the CH₄ anomaly decays exponentially with a perturbation lifetime of 12.5 years, matching that expected for the CH₄E simulation with a CH₄ total lifetime of 8.6 years and feedback factor F of 1.45, and similar to that found in previous studies (IPCC 1996; Prather 2007). We note that the decay of this perturbation is exponential and uniform after 2001, reflecting the principal natural mode in the tropospheric chemical scheme (Wild and Prather, 2000) and that it is largely unaffected by underlying changes in emissions or meteorology over the 2000–2017 period, as postulated by Holmes (2018). The impacts on CH₄ abundance are much smaller in the prescribed surface concentration (STD) run, as expected, but the changes in O₃ and OH persist over 2–3 years reflecting changes in short-lived chemical modes associated with CO–OH–O₃ interactions. In the emission-driven (CH₄E) run, the initial O₃ decrease in the first year is replaced by an O₃ increase from 2002 associated with the higher CH₄ abundance, and this decays with the same timescale as the CH₄ anomaly. While these changes are small, 1–2% of the O₃ change seen under the emission pulse in 2000, the accumulated burden change over the lifetime of the anomaly is 3.1 Tg·yr⁻¹, which offsets one third of the 9.2 Tg O₃ increase seen in 2000. Model studies using prescribed CH₄ concentrations are unable to represent these responses without additional adjustment, and this highlights the importance of using an emission-driven approach for assessing long-term climate responses.

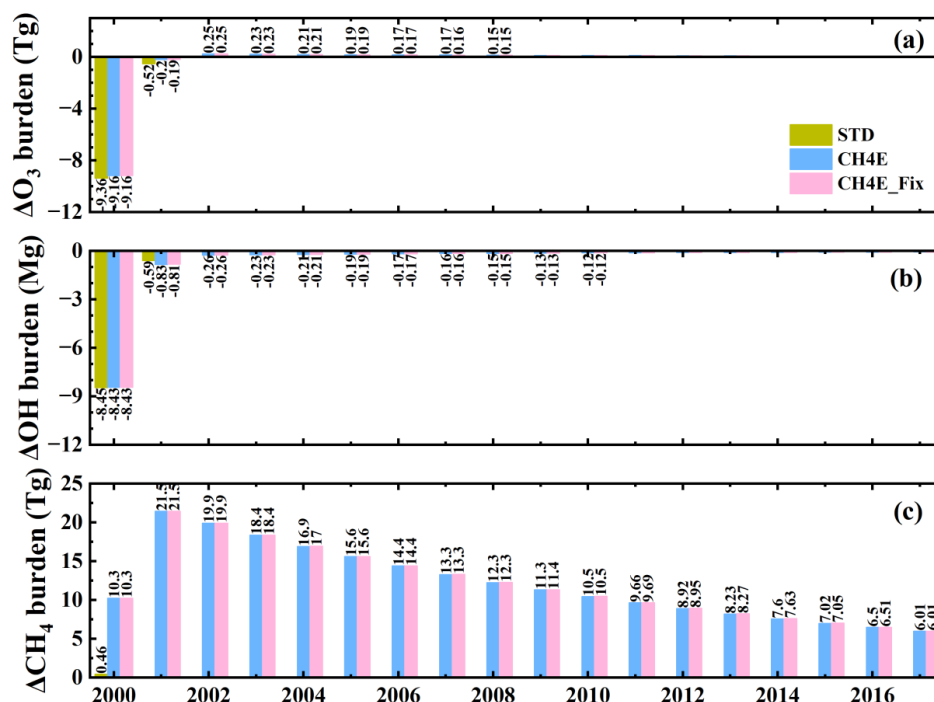


Figure 10. Temporal differences in (a) O_3 burden (Tg), (b) OH burden (Mg), and (c) CH_4 burden (Tg) from 2000 to 2017 in response to a 20 % reduction in NO surface emissions applied in year 2000 only. Results are shown for prescribed surface CH_4 concentration (STD) and emission-driven (CH4E) approaches, and for an emission-driven approach using repeating year 2000 precursor emissions and meteorology (CH4E_Fix).

4 Conclusions

A thorough assessment of how atmospheric CH_4 responds to its sources and sinks is essential for improving understanding of the CH_4 budget and advancing knowledge of air quality and anthropogenic climate change. In this study, we evaluated three treatments of CH_4 in a chemical transport model: prescribing mixing ratios throughout the troposphere, prescribing mixing ratios at the surface only, and incorporating a full treatment of CH_4 emissions. We investigated the impact of each approach on the simulated CH_4 budget and on how key tropospheric oxidants (O_3 and OH) respond to changing emissions and meteorology over the 2000-2017 period. We incorporate CH_4 emissions from anthropogenic, wetland, fires, geological, termite and biogenic sources which give a global CH_4 source of 602 Tg/yr over the 2000-2017 study period. We incorporate explicit treatments of surface uptake (averaging 35.6 Tg/yr), and stratospheric CH_4 loss (averaging 35.5 Tg/yr). The total atmospheric CH_4 lifetime in the emissions-driven simulation is 8.6 years, and the lifetime to chemical loss by reaction in the troposphere is 9.7 years. These modelled budget terms compare well with observation-based estimates from Saunio et al., (2025).

The emission-driven simulation successfully reproduces key features of the tropospheric CH_4 distribution seen in surface and aircraft CH_4 measurements, including the interhemispheric gradient, vertical distribution, seasonal variability, and long-term annual trends. As expected, it shows



substantially greater spatial variability in surface CH₄ than the prescribed concentration approaches, reflecting the spatial heterogeneity of source distributions. It also captures significantly stronger positive CH₄ trends in Africa, South America, India, and East and Southeast Asia, highlighting the importance of employing an emission-based framework to assess regional CH₄ trends.

- 520 The effect of different CH₄ treatments on tropospheric oxidants is relatively small. In the emissions-driven simulation surface O₃ is slightly higher globally due to the higher CH₄ levels, with the most pronounced increases (~0.4 ppbv) over East Asia, North America, and the southern hemisphere. The global tropospheric O₃ burden is 2.0 Tg (0.6%) larger than in the prescribed concentration simulation. Similarly, the OH burden is marginally (0.05%) greater. An increasing trend in surface O₃ is seen in
- 525 all model runs, notably over Europe, North America, South America, East Asia and South Asia. However, the emission-driven simulation shows a slower O₃ growth rate at southern hemisphere high latitudes and a faster growth rate in South and East Asia under the influence of high regional NO_x and VOCs concentrations. The emission-driven run shows a smaller positive trend in tropospheric OH due to enhanced OH consumption by CH₄ oxidation.
- 530 A 20% CH₄ perturbation results in a slightly larger increase in CH₄ burden in the emission-driven approach (~22%) than in the prescribed-concentration approaches (~20%). The conventional CH₄ feedback factors, which combine model-diagnosed OH loss with recommended lifetimes for other CH₄ sinks (Prather et al., 2012), are highly consistent across the different approaches (1.403–1.409), demonstrating that the simulated CH₄–OH feedback is largely insensitive to the choice of approach
- 535 taken. Feedback factors derived from model-diagnosed CH₄ sinks are slightly higher (~1.440), indicating sensitivity to the representation of CH₄ loss processes.

- The treatments of CH₄ have pronounced influences on the long-term response to changes in NO_x emissions. A 20% reduction in surface NO emissions produces similar immediate decreases in OH and O₃ across the different CH₄ treatments, but the emission-driven simulation additionally captures
- 540 a persistent CH₄ perturbation, with a peak burden increase of 21.5 Tg in the year following the perturbation and subsequently decaying with a perturbation lifetime of 12.5 years. This long-term CH₄ response is largely unaffected by interannual variations in meteorology and emissions of CH₄ and other species. The resulting increase in CH₄ subsequently enhances O₃, partially offsetting the initial O₃ decrease. These longer-lived CH₄–O₃ responses are not fully represented when CH₄
- 545 concentrations are prescribed, highlighting the importance of an emission-driven treatment of CH₄ for assessing the longer-term atmospheric response to emission perturbations.

- In summary, the updates to the FRS GC/UCI chemical transport model described here offer new capability and we have demonstrated that the model is capable of simulating CH₄ well when using emissions. While prescribed CH₄ concentrations may be sufficient for many studies of tropospheric composition, an emission-driven treatment is important for capturing longer-term CH₄–OH–O₃
- 550 responses to changes in emissions. This capability therefore provides a more complete framework for investigating the impacts of future emission changes on atmospheric chemistry and climate.

- 555 **Data availability.** The model output from the simulations described in this work are available on Zenodo (Hou et al., 2026; <https://zenodo.org/uploads/22030169>). CH₄ measurements from the NOAA Global Monitoring Laboratory network are publicly available at <https://gml.noaa.gov/ccgg/data/getdata.php?gas=ch4> (last access: 19 August 2026). Anthropogenic CH₄ emissions from the CMIP7 input4MIPs historical forcing dataset are available through ESGF at



560 <https://esgf-node.ornl.gov/search/input4mips/> (last access: 19 August 2026). Wetland emissions from the WetCHARTs v1.0 extended ensemble are available at <https://doi.org/10.3334/ORNLDAAAC/1502>. Fire emissions from GFED4s are available at <https://www.globalfiredata.org/> (last access: 19 August 2026). Biogenic CH₄ emissions from CAMS-GLOB-BIO v3.1 and termite emissions from CAMS-GLOB-TERM v1.1 are available through the Copernicus Atmosphere Data Store at
 565 <https://ads.atmosphere.copernicus.eu/datasets/cams-global-emission-inventories> (last access: 19 August 2026). Geological CH₄ emissions from the Global Carbon Project Methane Budget 2024 dataset are available at <https://doi.org/10.18160/GKQ9-2RHT> (Martinez et al., 2024).

Author contributions. XH and RH designed the study and updated the emission inventories. XH
 570 and OW updated the model code. XH conducted the modeling and data analysis with contributions from RH, OW, JL, and YW. JL and YW discussed the results and offered valuable comments. XH, RH and OW wrote the paper with input from all authors.

Competing interests. The authors declare that they have no conflict of interest.

575
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