Chemistry Aerosol Working Group (CAWG) meeting
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Assessing the drivers of the temporal variations in methane loss

Benjamin Gaubert[1], M. Amin Mirrezaei[2], Shuo Chen[3] I. Ortega[1] K. McKain[4], L. Bruhwiler[4], Y. Oh[4,5], A. Saiz-Lopez[6], R. Fernandez[7,8], L. Emmons[1], C. Feng[9], Y. Xu[9] and A. F. Arellano[2]

[1]Atmospheric Chemistry Observations Modeling Laboratory (ACOM), NSF NCAR; [2]University of Arizona; [3]Purdue University; [4]Global Monitoring Laboratory, NOAA; [5]CIRES, University of Colorado; [6]Institute of Physical Chemistry Blas Cabrera, CSIC; [7]ICB-CONICET; [9]Texas A&M University

The red emissions are characteristic spectral emissions from excited atomic oxygen (O^1D) in the thermosphere, here during a solar storm near NSF NCAR foothills laboratory.

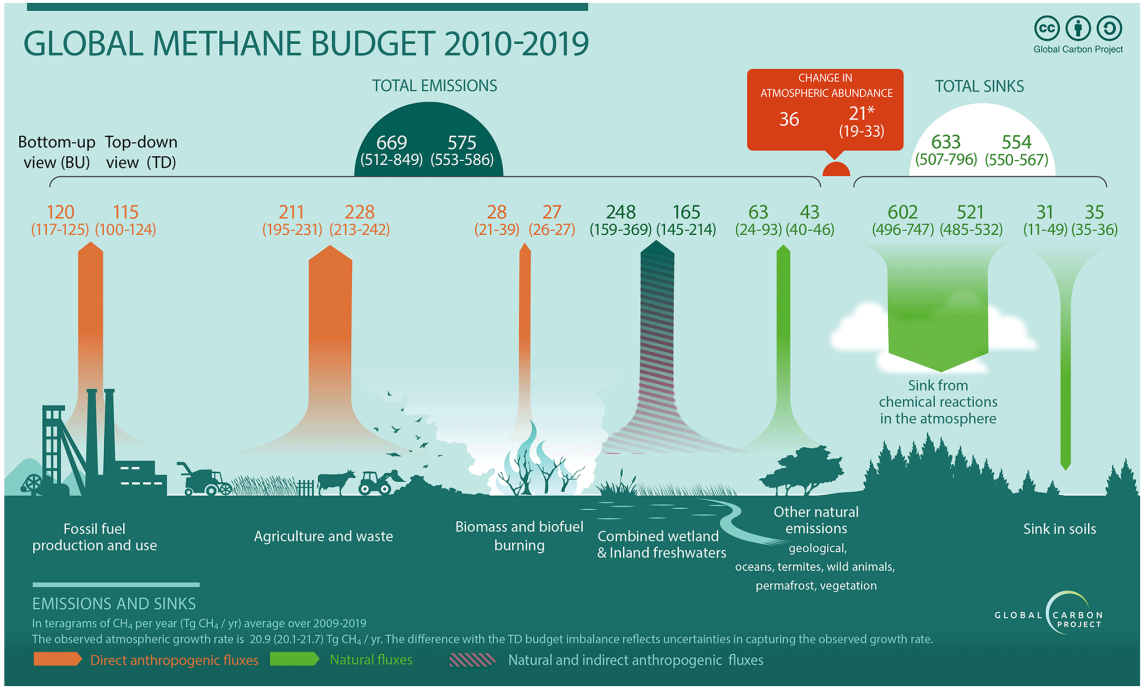
CH_4 budget

Sources:

- 65 % of the emissions comes from direct anthropogenic sources (fossil fuel, agriculture, waste).
- Combined wetland and inland freshwater emissions are the largest sources of methane

Losses:

- 90 % of the loss is its oxidation by OH radicals.



Saunois et al., (2025); Global Methane Budget 2000–2020

Trends in atmospheric CH_4 mole fractions

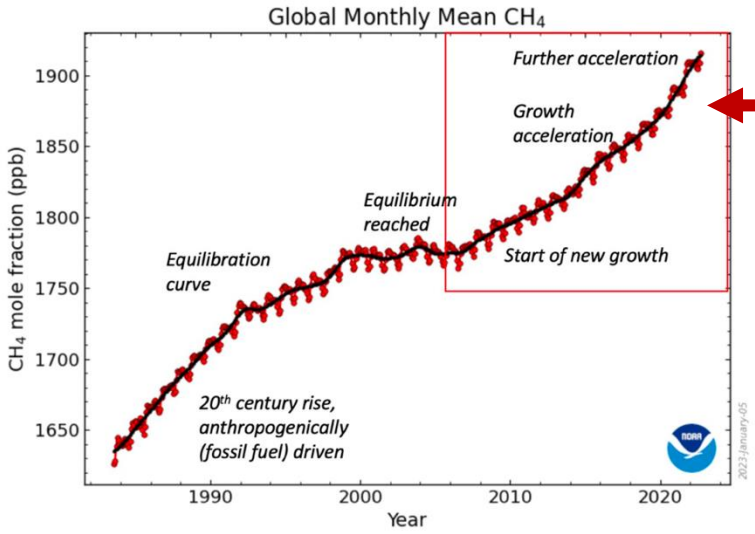


Figure 1. Global monthly mean of atmospheric CH_4, NOAA network. Note convex equilibration curve to 2006; concave acceleration curve from 2007. The convex curve from the 1980s to early 2000s is consistent with relaxation to a source-sink steady state (Dlugokencky et al., 2003), but then the start of a renewed growth became apparent in 2007, and the slope of the new concave curve steepened in 2013 (acceleration of growth), followed by further acceleration in 2020. Modified from Lan et al. (2022). Data from https://gml.noaa.gov/dv/data.html.

Nisbet et al. (2023):
"Recent studies point to strongly increased emissions from wetlands, especially in the tropics"

- Large tropical source from wetland emissions, and agriculture.
- Possible contribution by negative anomalies in OH radicals (e.g., Chen et al.., 2025) driven by NOx emissions reduction following COVID-19 lockdowns and extreme fire emissions (e.g., Australia 2019)

Nisbet, E. G., et al. (2023), Atmospheric Methane: Comparison Between Methane's Record in 2006–2022 and During Glacial Terminations *Global Biogeochemical Cycles*.

Chen et al., (2025), Converging evidence for reduced global atmospheric oxidation in 2020, *National Science Review*.

Hydroxyl radical ·OH

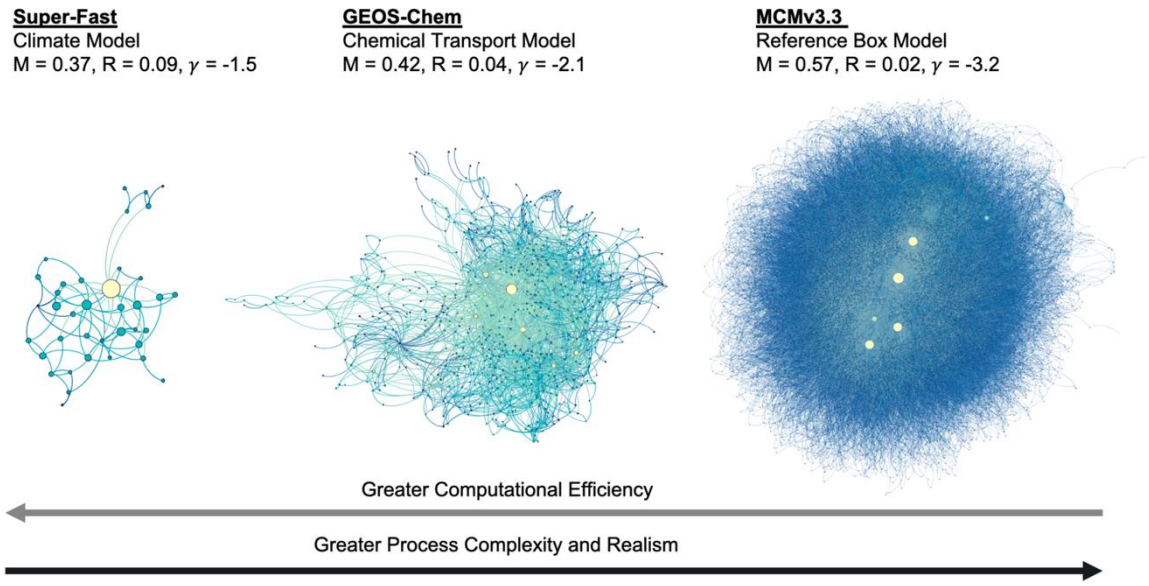


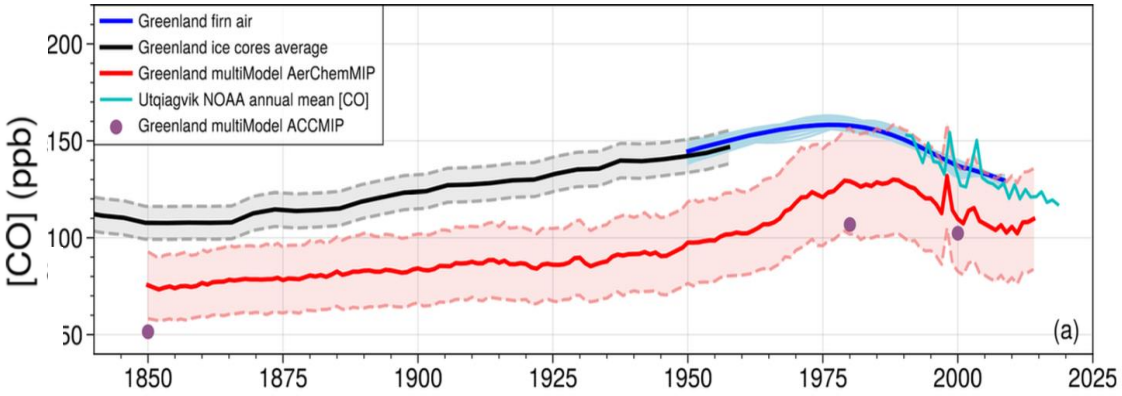
Figure 2. A visual representation of the species-reaction graphs used in this work, analogous to Figure 1. Vertex color (dark-light) and size (small-large) scales on a per-graph basis with the out-degree centrality (the number of edges leaving a node, see Section 5). Use cases and global graph properties (M = modularity, R = Reciprocity, γ = Gamma fitting parameter, see Section 4) are summarized below graph labels.

Vertex centrality: OH is the most important species of atmospheric chemistry

Silva, S. J., et al., A Graph Theoretical Intercomparison of Atmospheric Chemical Mechanisms, *Geophysical Research Letters*, 2021.

Reconciliate atmospheric OH estimates

- Uncertainties of 10 % in methyl-chloroform inversions (e.g., global methane budget 2025).
- chemistry-climate model (CCM) prognostic OH have uncertainties of around 20 %
- CCMs tend to **overestimate the OH distribution**
- Current estimates diverge on the **latitudinal and temporal distribution of OH**



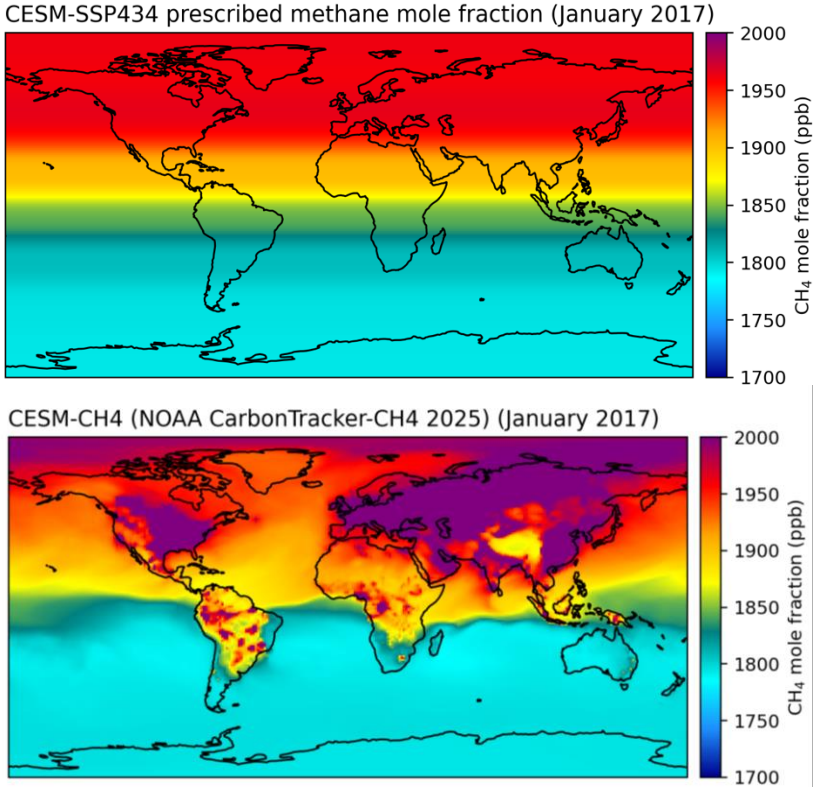
"The AerChemMIP model mean is about 20% lower than ice archive datasets at northern high latitudes during the entire 1850–2014 period."

Faïn, X., et al., (2025)
Saunois et al., (2025)

Emission-driven methane simulations:

- Evaluating the impact of chemistry and chemistry changes on the methane growth rate.
- Include and quantify chemical feedback.
- Feedback on emissions processes within the Earth System (e.g., soil, fires, and wetlands response to climate)

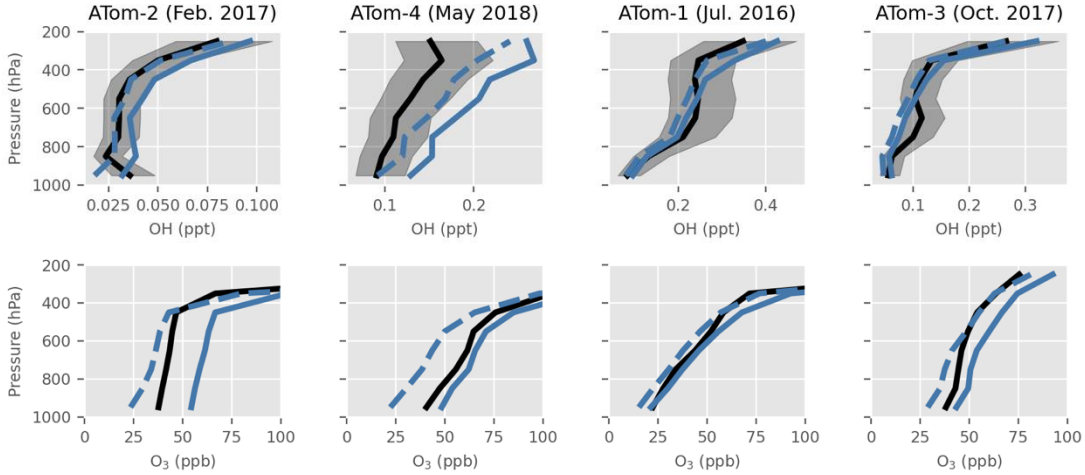
CESM-CH4: methane flux emission-driven simulation
Methane posterior flux are provided by NOAA CarbonTracker-CH4 2025

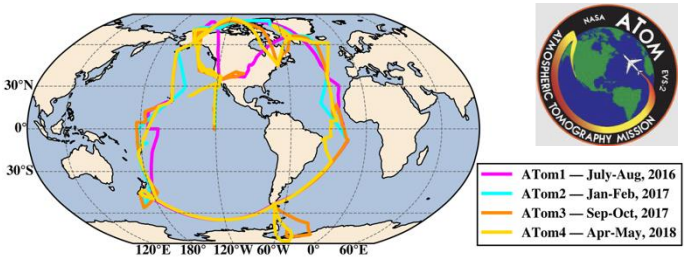


Impact of short-lived halogen (SLH) chemistry

CESM2.2 SLH chemistry (Fernandez et al., 2025)

Simulations	Chemistry	Methane	Anthro CO emis.	Fire CO emis.
TS1.2-CMIP6	TS1.2	prescribed (CMIP6)	CAMS-GLOB-ANT_v5.3	FINN2.5
SLH-CMIP6	**TS1.2-SLH**	prescribed (CMIP6)	CAMS-GLOB-ANT_v5.3	FINN2.5





	Bias over Pacific Ocean >30°N		Bias over Atlantic Ocean >30°N	
Simulation	O_3 (ppb)	OH (ppq)	O_3 (ppb)	OH (ppq)
TS1.2-CMIP6	14.6	28.2	16.6	27.6
SLH-CMIP6	-1.5	3.6	2.3	23.5

Fernandez et al., (2025). EGUsphere
Mirrezaei, et al. (2026). Toward realistic prognostic modeling of the methane chemical loss. JGR

Tropospheric Ozone burden vs OH

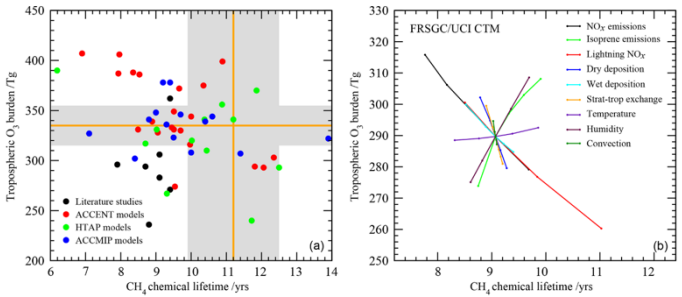
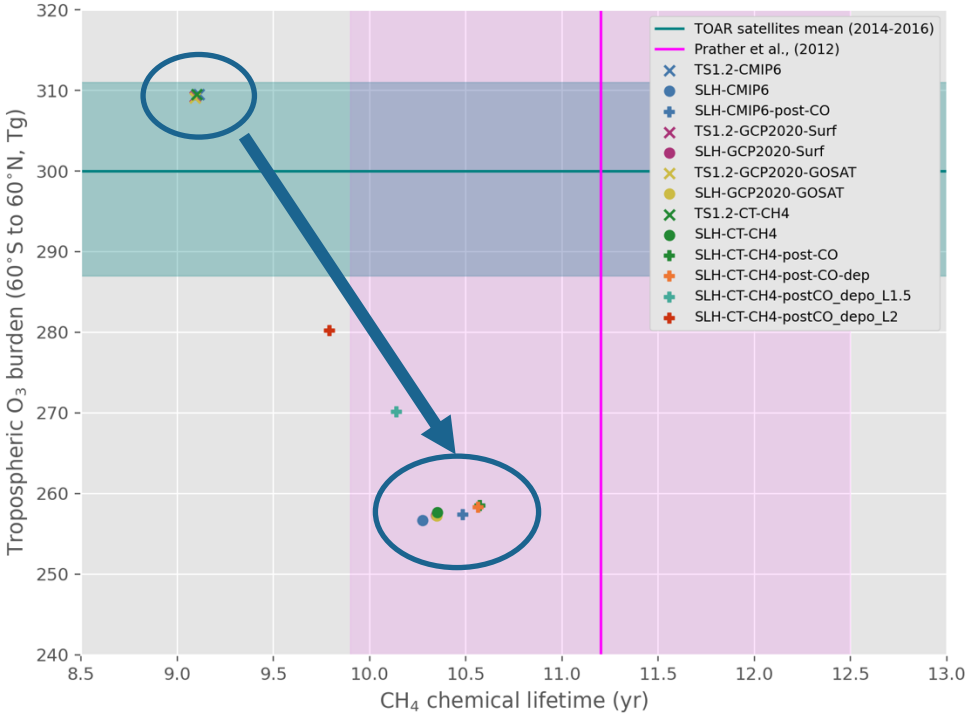


Figure 1. Tropospheric oxidant budgets from previous published studies and model intercomparisons **(a)**, along with measurement-based estimates of the tropospheric O_3 burden and CH_4 lifetime (shaded regions). Panel **(b)** shows results from one-at-a-time sensitivity studies with a single model revealing the extent to which individual processes can influence the budgets (see Wild, 2007, for details). Note that results in **(a)** represent differing emissions and meteorological years (study details are given in Table 1) and that **(b)** covers only part of the parameter space shown in **(a)**.

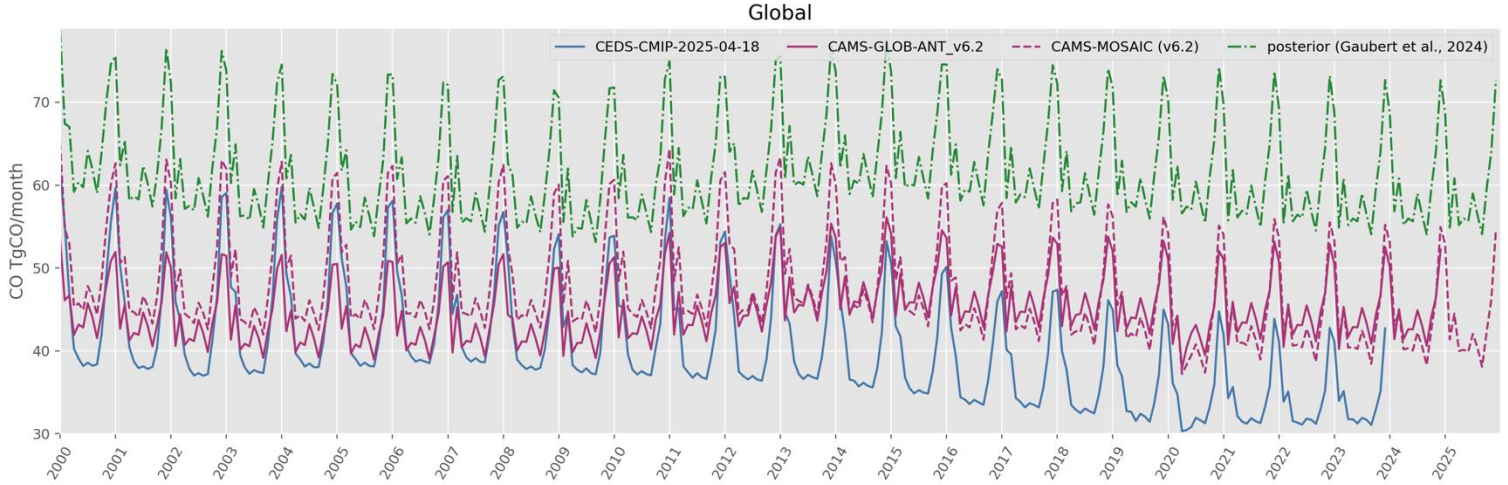
Table 1. Global tropospheric metrics from previous model studies.

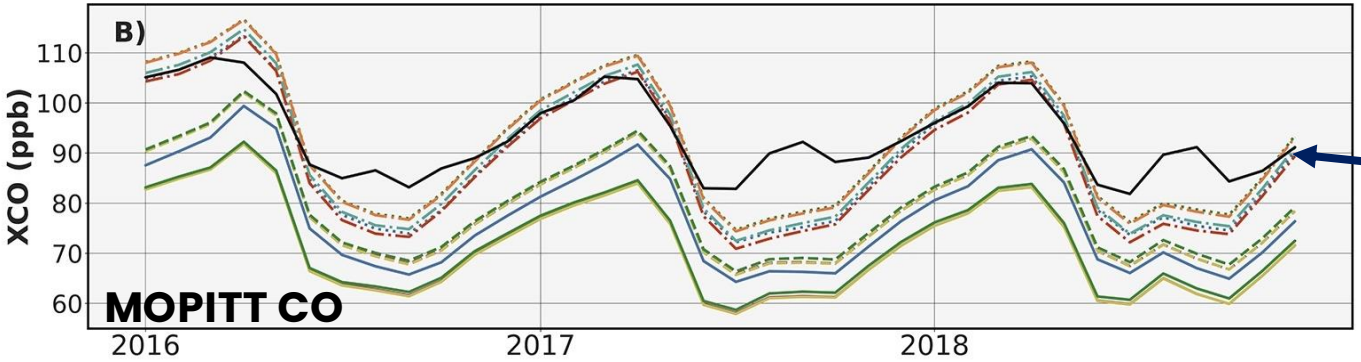
Studies	Number	O_3 burden	CH_4 lifetime	References
Early literature studies	33 studies	307 ± 38 Tg		Wild (2007)
ACCENT intercomparison	21 models	344 ± 39 Tg	9.6 ± 1.4 years	Stevenson et al. (2006)
HTAP intercomparison	12 models	328 ± 41 Tg	10.2 ± 1.7 years	Fiore et al. (2009)
ACCMIP intercomparison	14 models	337 ± 23 Tg	9.8 ± 1.6 years	Young et al. (2013), Voulgarakis et al. (2013)
Observational estimates		335 ± 20 Tg	11.2 ± 1.3 years	Wild (2007), Prather et al. (2012)



Wild et al., Global sensitivity analysis of chemistry–climate model budgets of tropospheric ozone and OH: exploring model diversity, 2020

Addressing OH biases: impact of CO emissions





SLH chemistry with posterior CO emissions

CESM2.2/CAM-chem simulations

Simulation	CH_4 Emiss.	Anthro Emiss.	Fire Emiss.	Anthro CO Emiss.
CESM	prescribed methane (SSP434 after 2015)	CAMS-MOSAIC	FINN2.5	MOPITT inversion**
CESM-245	prescribed methane (SSP245 after 2015)	CAMS-MOSAIC	FINN2.5	MOPITT inversion**
CESM-CH4	NOAA CT-CH4 2025*	CAMS-MOSAIC	FINN2.5	MOPITT inversion**

- 2002 as spinup, 2003 to 2022 included

f09, 32 vertical layers, and nudged to MERRA-2
*NOAA CarbonTracker-CH4 2025 (Oh et al., 2025)
** From Gaubert et al., (2024)

Oh, Y., et al.,. CarbonTracker CH_4 2025.
NOAA Global Monitoring Laboratory, 2025.
Gaubert, et al. "Nonlinear and non-Gaussian ensemble assimilation of MOPITT CO." JGR: Atmospheres, 2024.

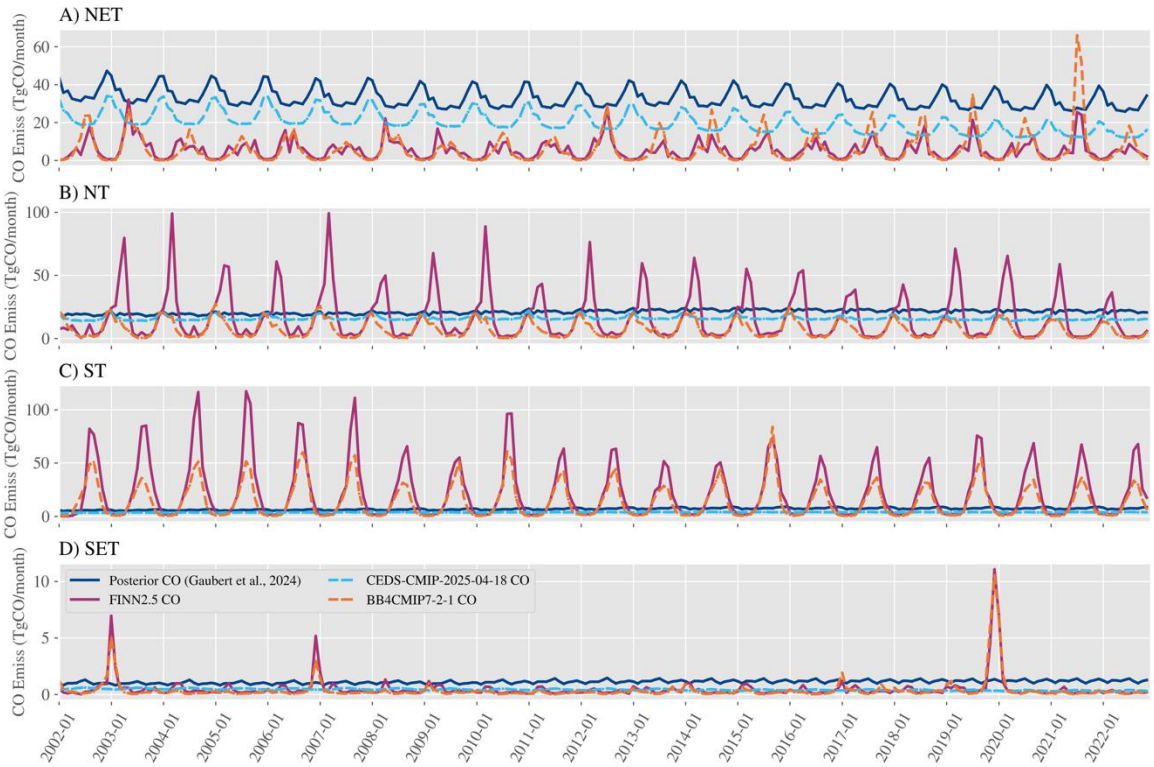
CESM2.2/CAM-chem simulations

Simulation	CH_4 Emiss.	Anthro Emiss.	Fire Emiss.	Anthro CO Emiss.
CESM	prescribed methane (SSP434 after 2015)	CAMS-MOSAIC	FINN2.5	MOPITT inversion**
CESM-245	prescribed methane (SSP245 after 2015)	CAMS-MOSAIC	FINN2.5	MOPITT inversion**
CESM-CH4	NOAA CT-CH4 2025*	CAMS-MOSAIC	FINN2.5	MOPITT inversion**
CESM-CH4-CMIP7	NOAA CT-CH4 2025*	CEDS-CMIP-2025-04-18	BB4CMIP7	CEDS-CMIP-2025-04-18
CESM-CH4-CMIP7-CO	NOAA CT-CH4 2025*	CEDS-CMIP-2025-04-18	BB4CMIP7	CEDS-CMIP-2025-04-18 x 1.3
CESM-CH4-2003	NOAA CT-CH4 2025* (2003 emissions)	CAMS-MOSAIC	FINN2.5	MOPITT inversion**

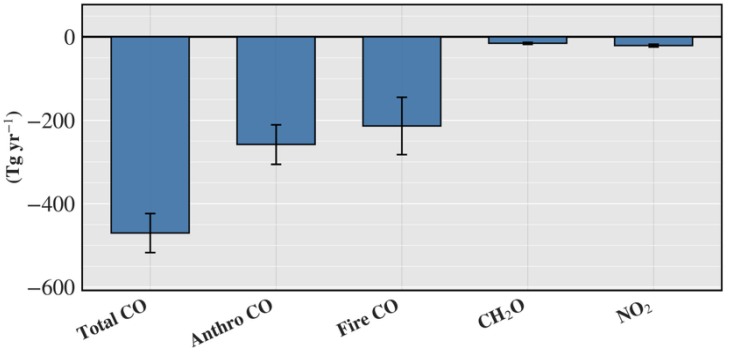
f09, 32 vertical layers, and nudged to MERRA-2
*NOAA CarbonTracker-CH4 2025 (Oh et al., 2025)
** From Gaubert et al., (2024)

Oh, Y., et al.,. CarbonTracker CH_4 2025.
NOAA Global Monitoring Laboratory, 2025.
Gaubert, et al. "Nonlinear and non-Gaussian ensemble assimilation of MOPITT CO." JGR: Atmospheres, 2024.

CMIP7 CO emissions:



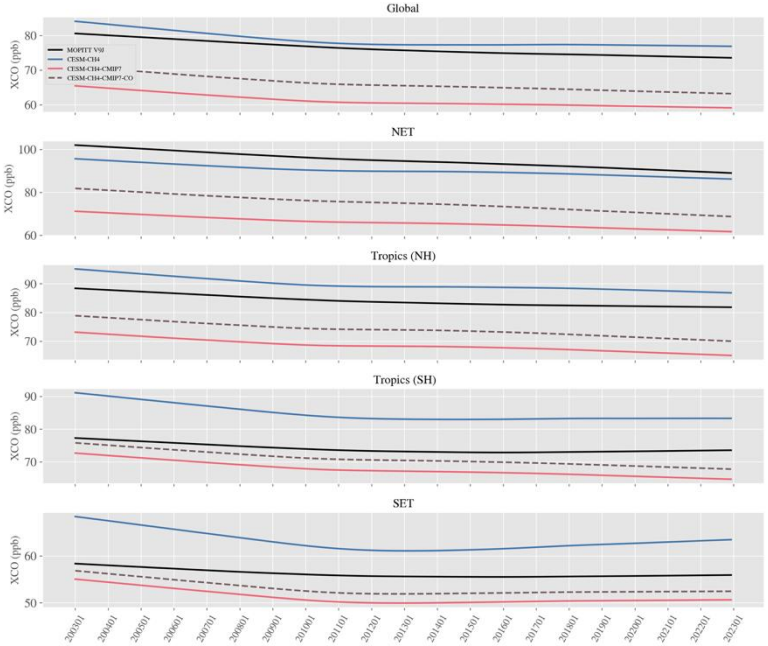
CESM-CH4-CMIP7 - CESM-CH4



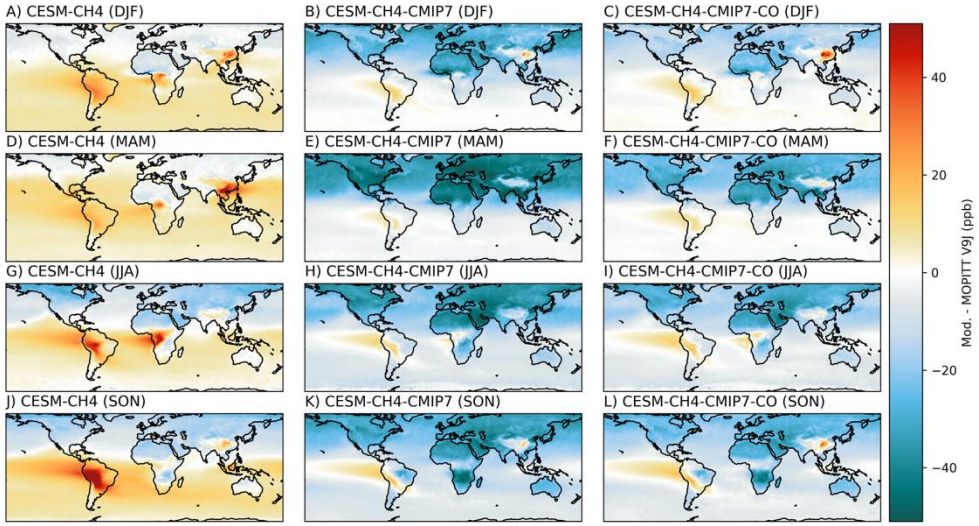
FINN2.5 estimates higher fire CO emission across NT, ST and SET

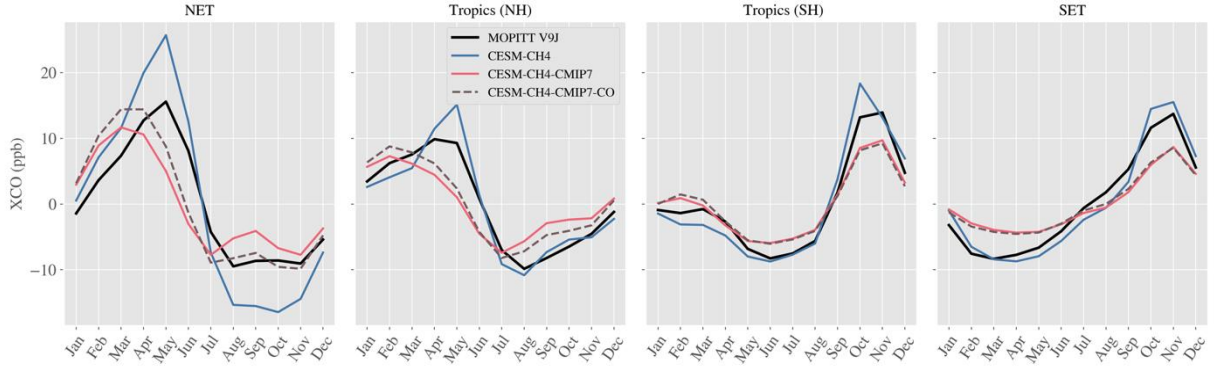
Posterior CO has higher value compared to CEDS-CMIP

Comparison with MOPITT V9J CO

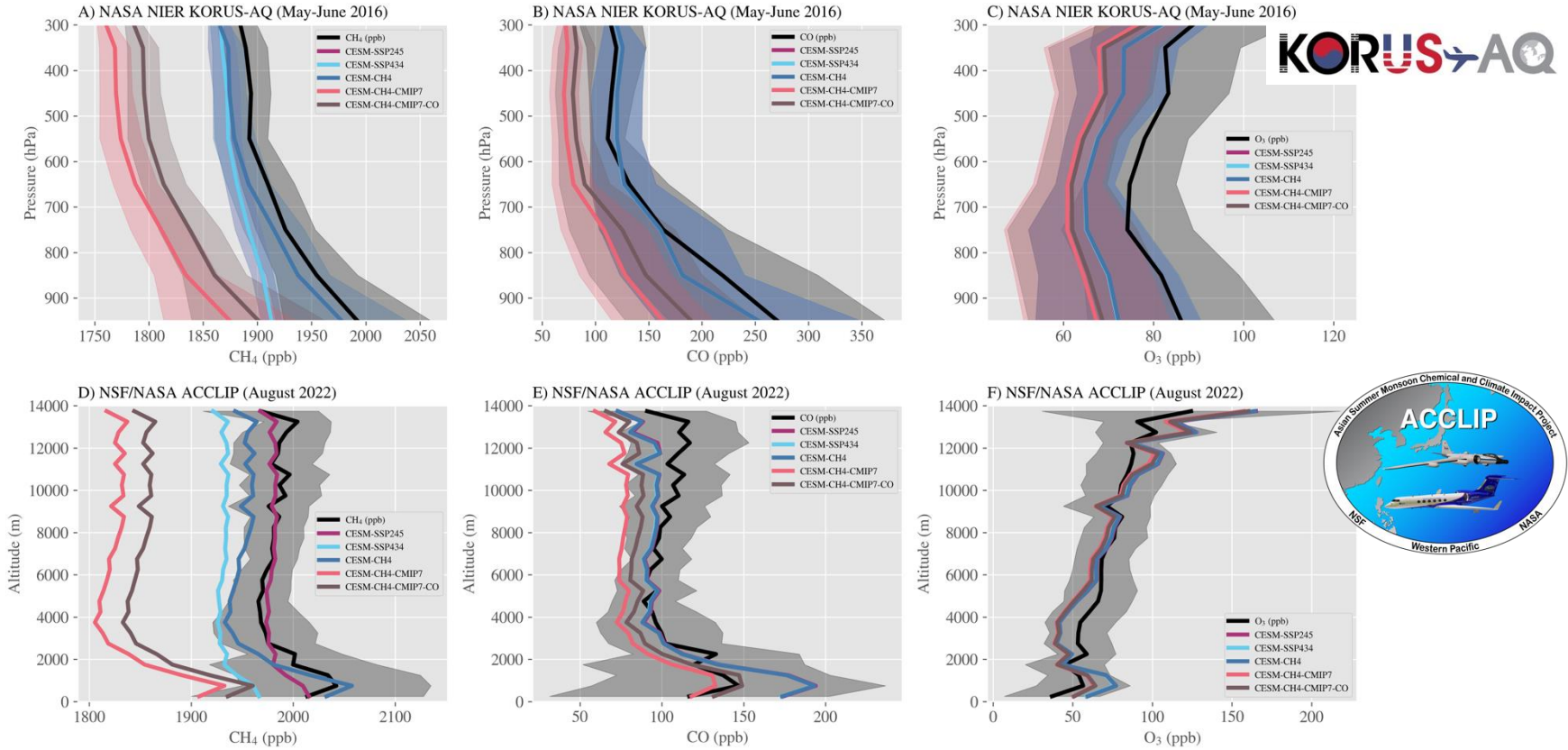


Comparison with MOPITT V9J CO



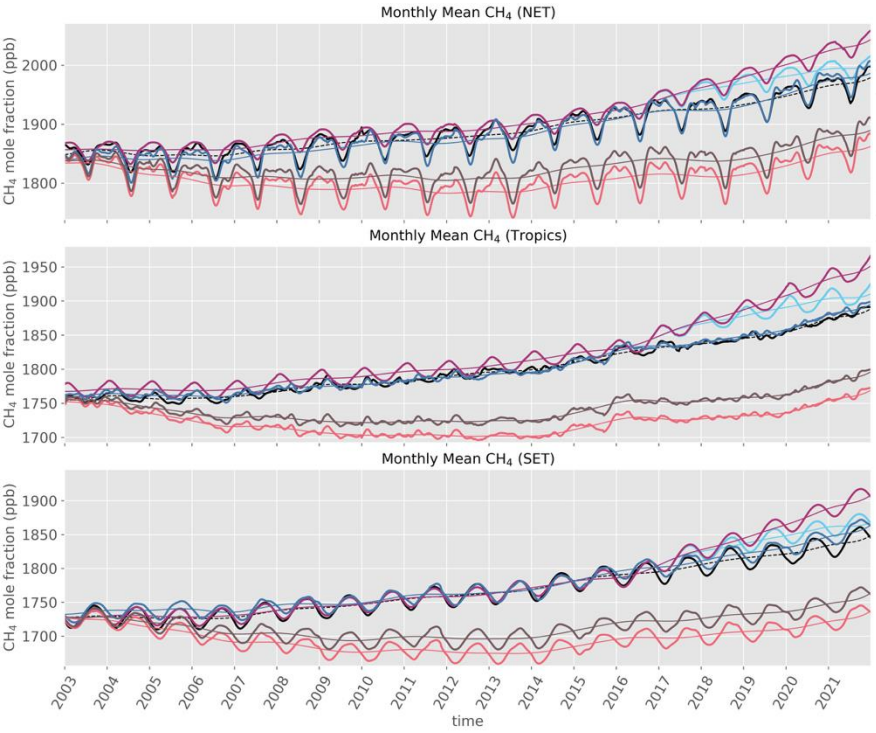


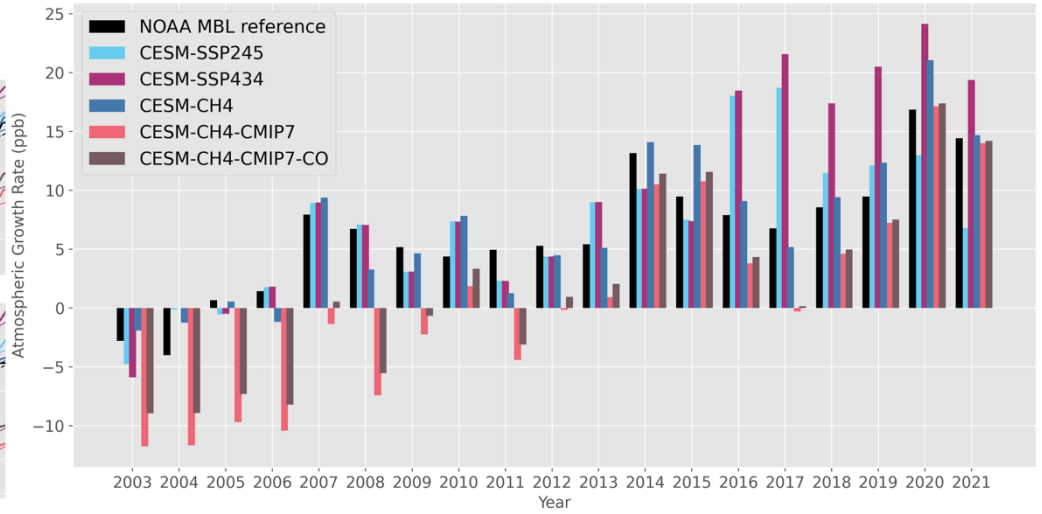
Comparison with Airborne field campaign



CESM-CH4-CMIP7 shows a persistent low bias throughout the troposphere.
CESM-CH4 reproduces observed vertical gradients for CH_4, CO, and O_3

NOAA Marine Boundary Layer Atmospheric Growth Rate



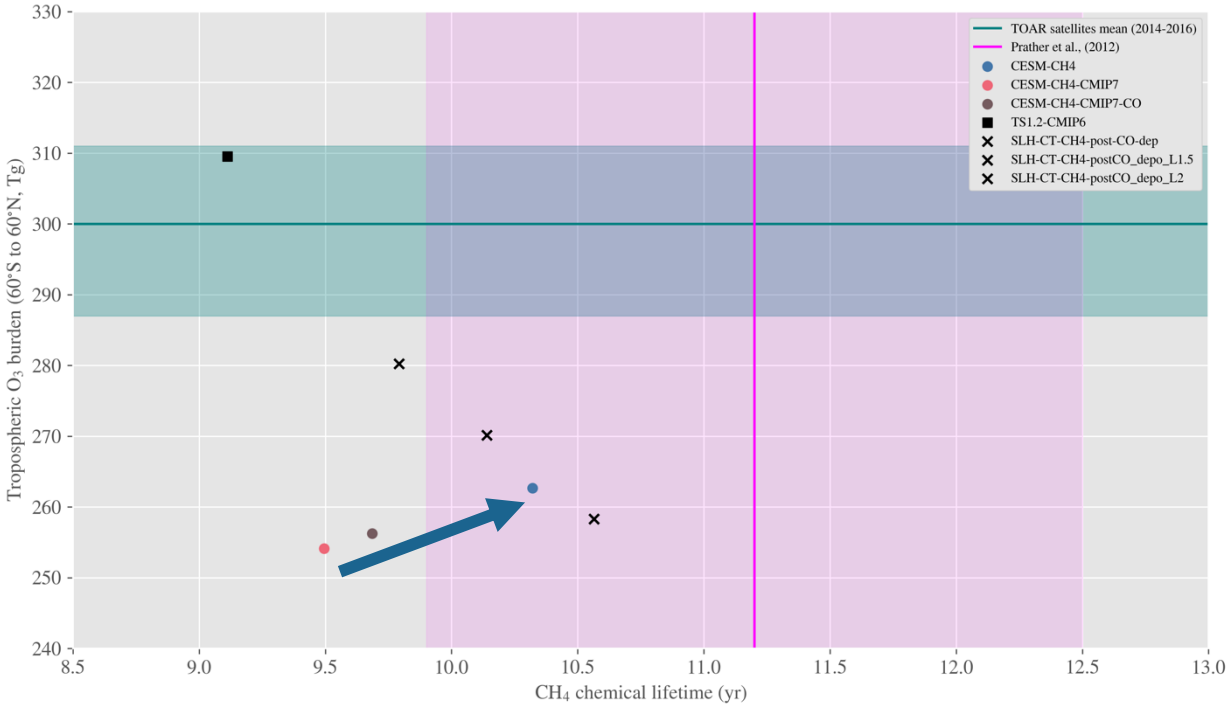


CESM: observation-driven prescribed methane is slightly higher than the NOAA MBL data, while scenarios after 2015 overestimate methane

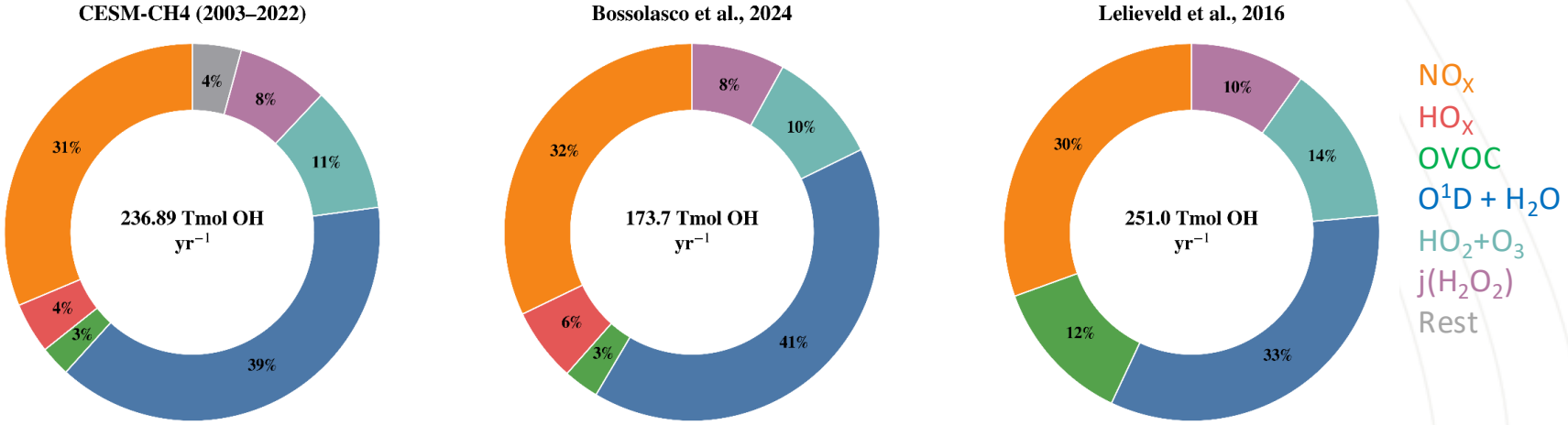
Modelled Atmospheric Growth Rate Both is better in emission-driven simulations than in observation-driven prescribed field

Low tropical CO emissions prevents correct CH4 simulation with CMIP7 forcings

Tropospheric Ozone burden vs OH

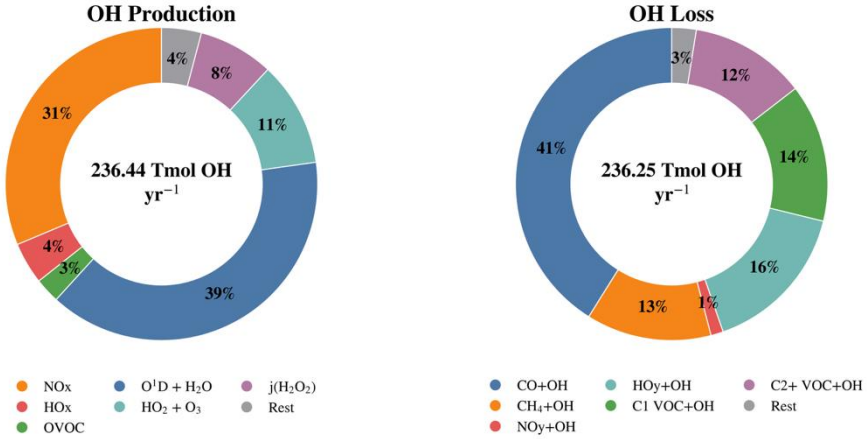


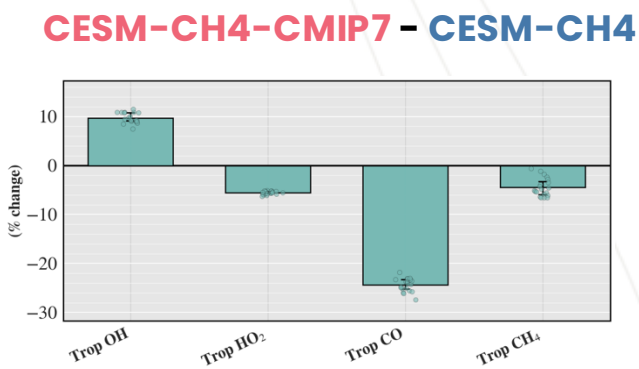
OH budget (2010–2019)

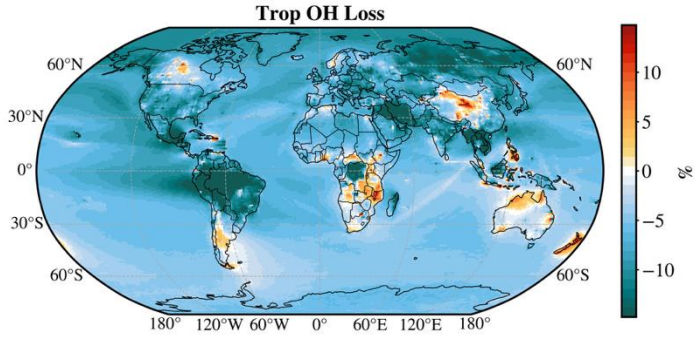


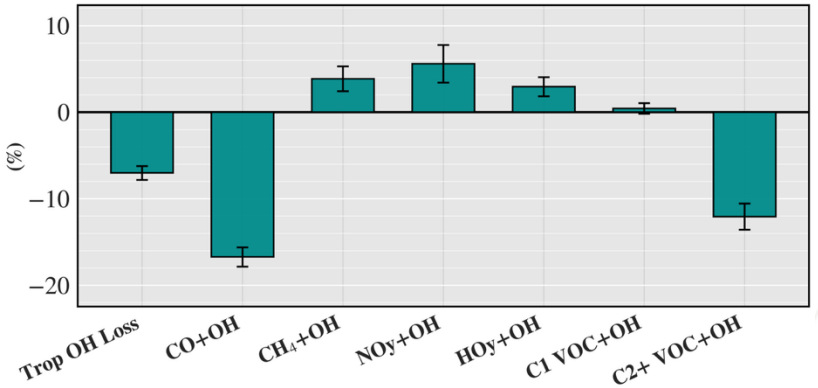
- OH recycling probability of 0.61: global and annual OH is well buffered to perturbations

OH budget (2010-2019)





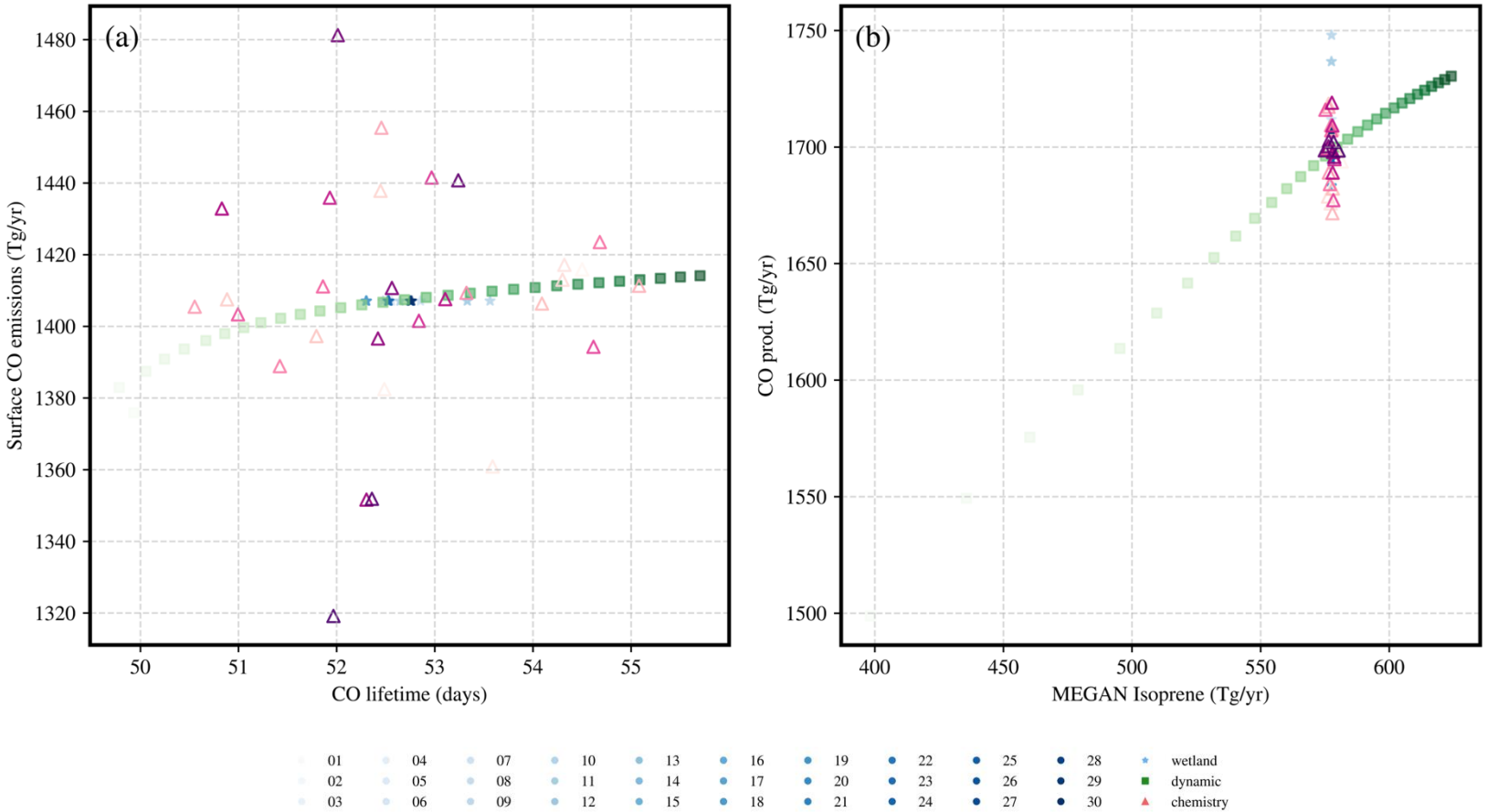




Ensemble Simulations: Nudging to ERA5 experiment

nens	U	V	T	Q
1	0.1	0.1	0	0
2	0.11	0.11	0.01	0.01
3	0.12	0.12	0.02	0.02
4	0.13	0.13	0.03	0.03
5	0.14	0.14	0.04	0.04
6	0.15	0.15	0.05	0.05
7	0.16	0.16	0.06	0.06
8	0.17	0.17	0.07	0.07
9	0.18	0.18	0.08	0.08
10	0.19	0.19	0.09	0.09
11	0.2	0.2	0.1	0.1
12	0.21	0.21	0.11	0.11
13	0.22	0.22	0.12	0.12
14	0.23	0.23	0.13	0.13
15	0.24	0.24	0.14	0.14
16	0.25	0.25	0.15	0.15
17	0.26	0.26	0.16	0.16
18	0.27	0.27	0.17	0.17
19	0.28	0.28	0.18	0.18
20	0.29	0.29	0.19	0.19
21	0.3	0.3	0.2	0.2
22	0.31	0.31	0.21	0.21
23	0.32	0.32	0.22	0.22
24	0.33	0.33	0.23	0.23
25	0.34	0.34	0.24	0.24
26	0.35	0.35	0.25	0.25
27	0.36	0.36	0.26	0.26
28	0.37	0.37	0.27	0.27
29	0.38	0.38	0.28	0.28
30	0.39	0.39	0.29	0.29

Ensemble Simulations



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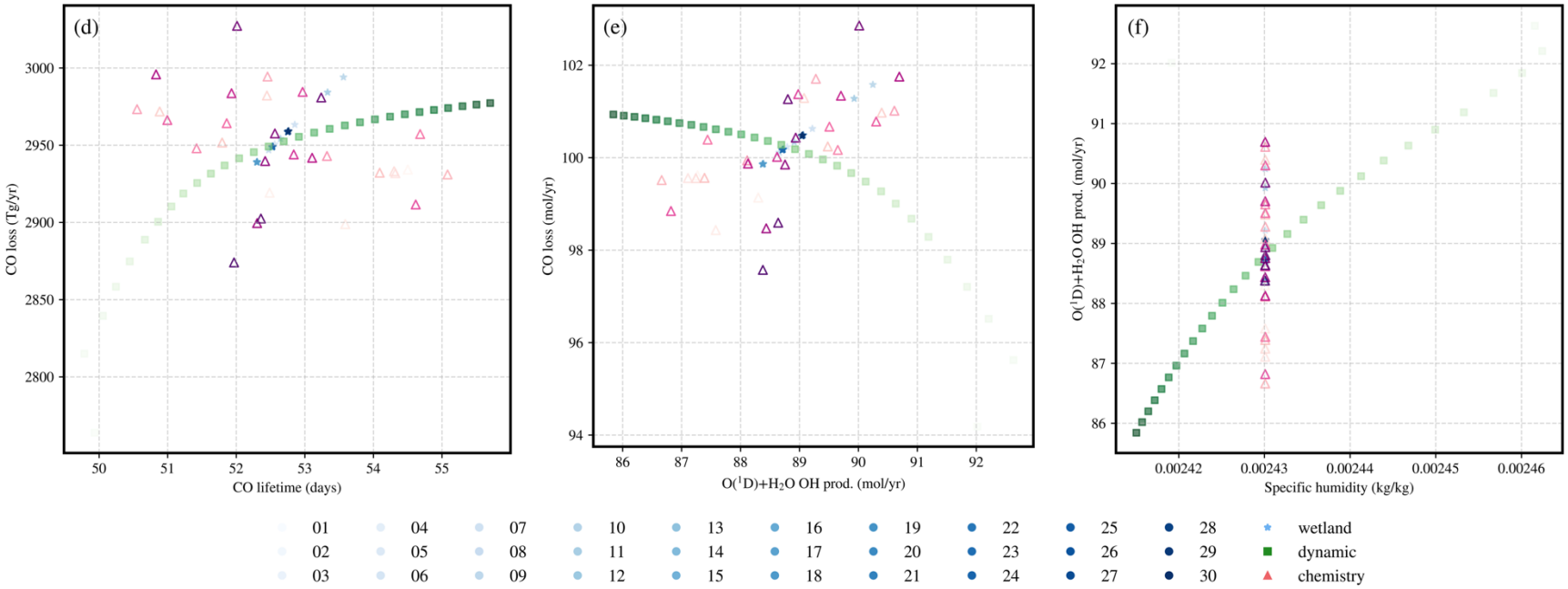

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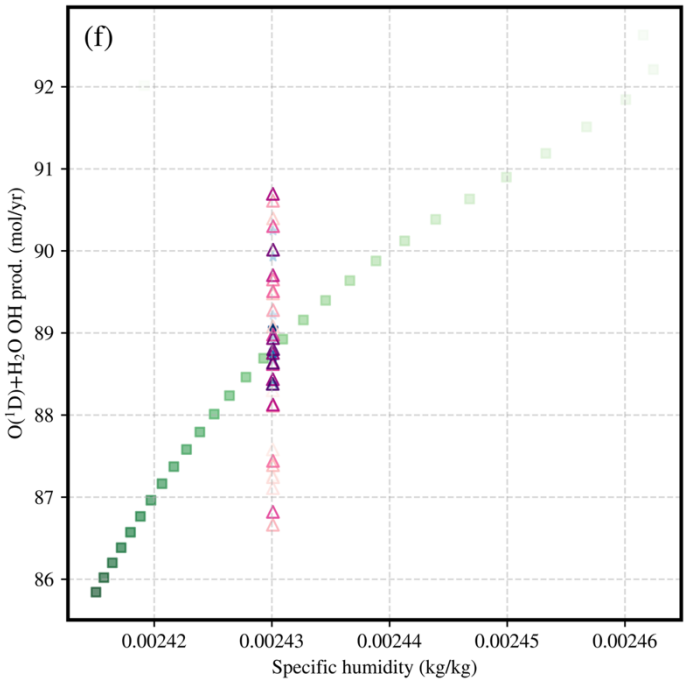
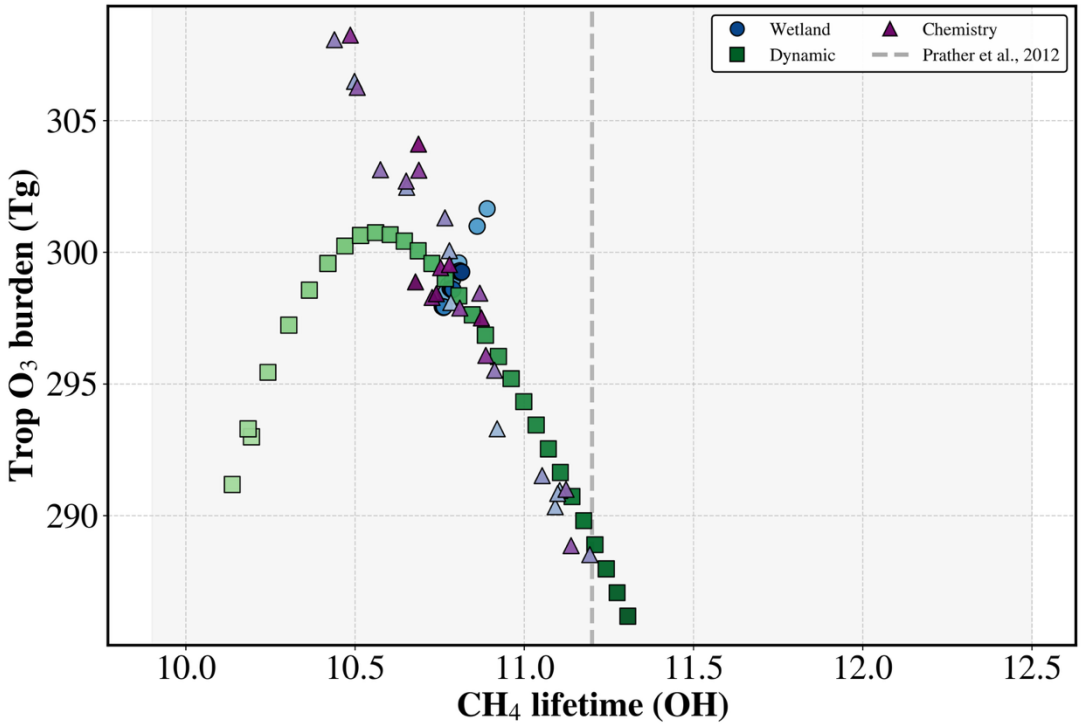




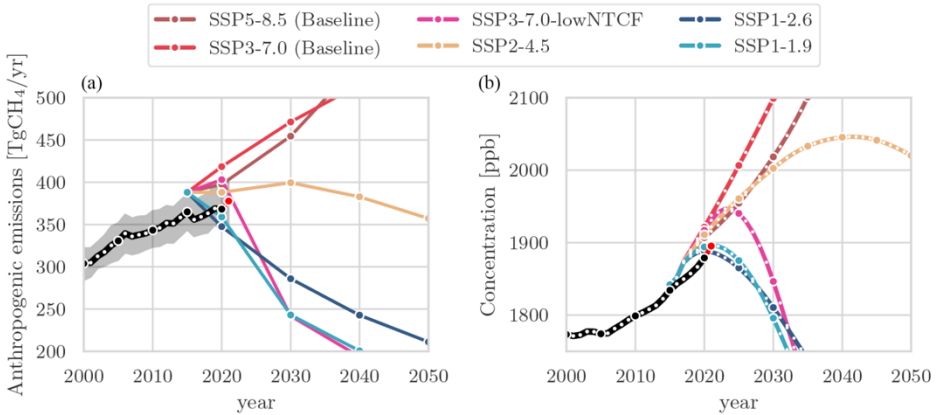
Conclusions

- Including halogen chemistry reduces global tropospheric OH, increasing the CH4 lifetime by ~1 year.
- Emission-driven CH4 simulations show sensitivity of OH and CH4 to anthropogenic and fire CO emissions, consistent with satellite and aircraft constraints.

- CESM-CH4 reproduces more realistically MOPITT CO, surface in-situ CH4, and aircraft in-situ CO, CH_4 and O_3 observations compared to CMIP7-based input emissions.

- Modelled Atmospheric Growth Rate Both is better in one emission-driven simulations than in the simulations using observation-driven prescribed fields.

- Stronger nudging to ERA-5 provides higher emissions from vegetation (MEGAN) and lower ozone photolysis and source of OH, changing the lifetime by ~1 year.



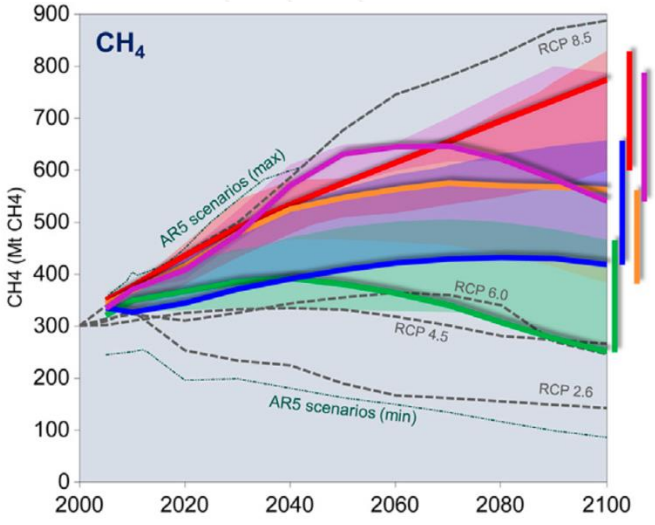


Figure 2. **(a)** Global anthropogenic methane emissions (including biomass burning) over 2000–2050 from historical inventories (black line and shaded grey area) and future projections (coloured lines) (in Tg CH_4 yr^{-1}) from selected scenarios harmonised with historical emissions (CEDS) for CMIP6 activities (Gidden et al., 2019). Historical mean emissions correspond to the average of anthropogenic inventories listed in Table 1 added to the GFEDv4.1s (van der Werf et al., 2017) biomass burning historical emissions. **(b)** Global atmospheric methane concentrations for NOAA surface site observations (black) and projections based on SSPs (Riahi et al., 2017) with concentrations estimated using MAGICC (Meinshausen et al., 2017, 2020). Red dots show the last year available (2022 for observations).

Ensemble Simulations