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Investigating the role of chemistry on the methane budget in emission-driven CH_4 simulations

Chemistry-Climate Working Group, CESM Workshop 2025

Tuesday, June 10, 2025

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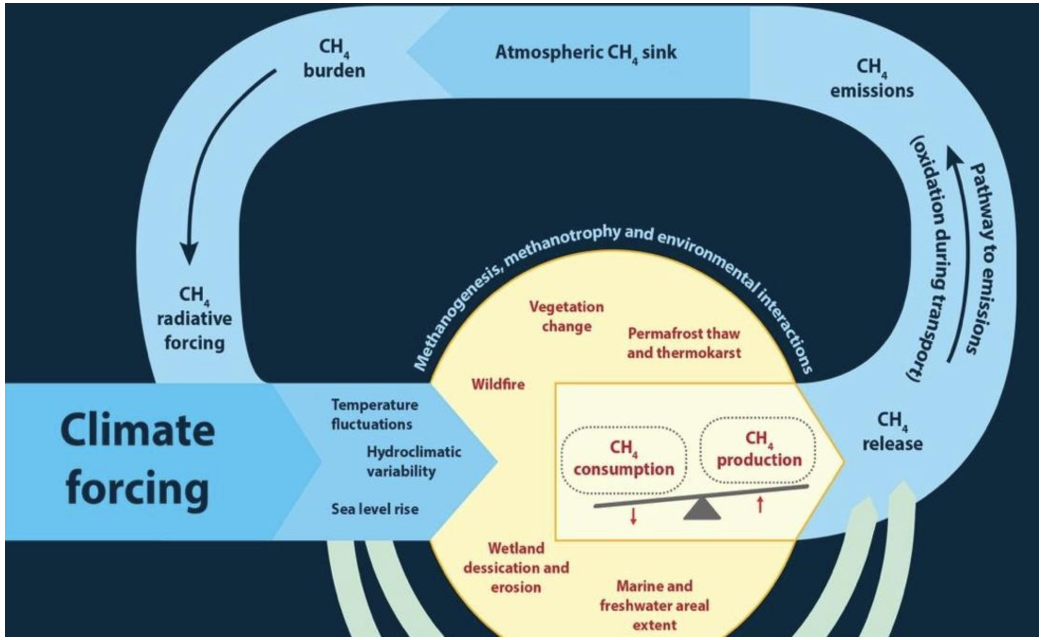
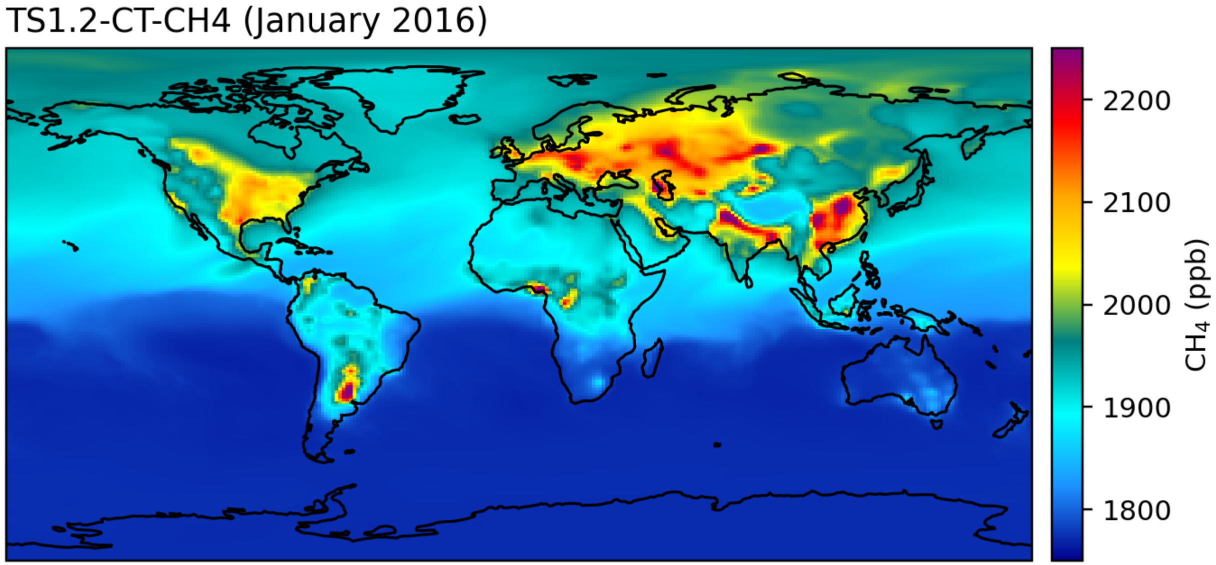
Emission-driven methane simulations: emissions-chemistry-climate feedback





- ✓ Understand the methane budget sources and sinks
- ✓ Evaluating the impact of chemistry and chemistry changes on the methane growth rate
- ✓ Include chemical feedback
- ✓ include further climate feedback emissions processes (e.g., soil and wetlands response to climate)

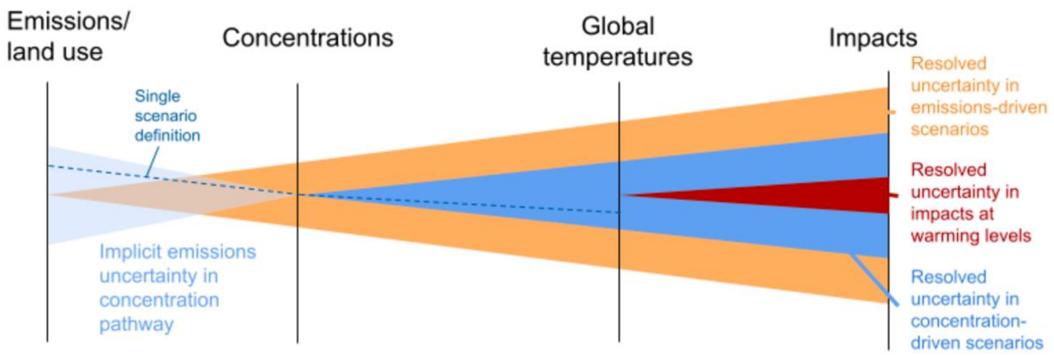
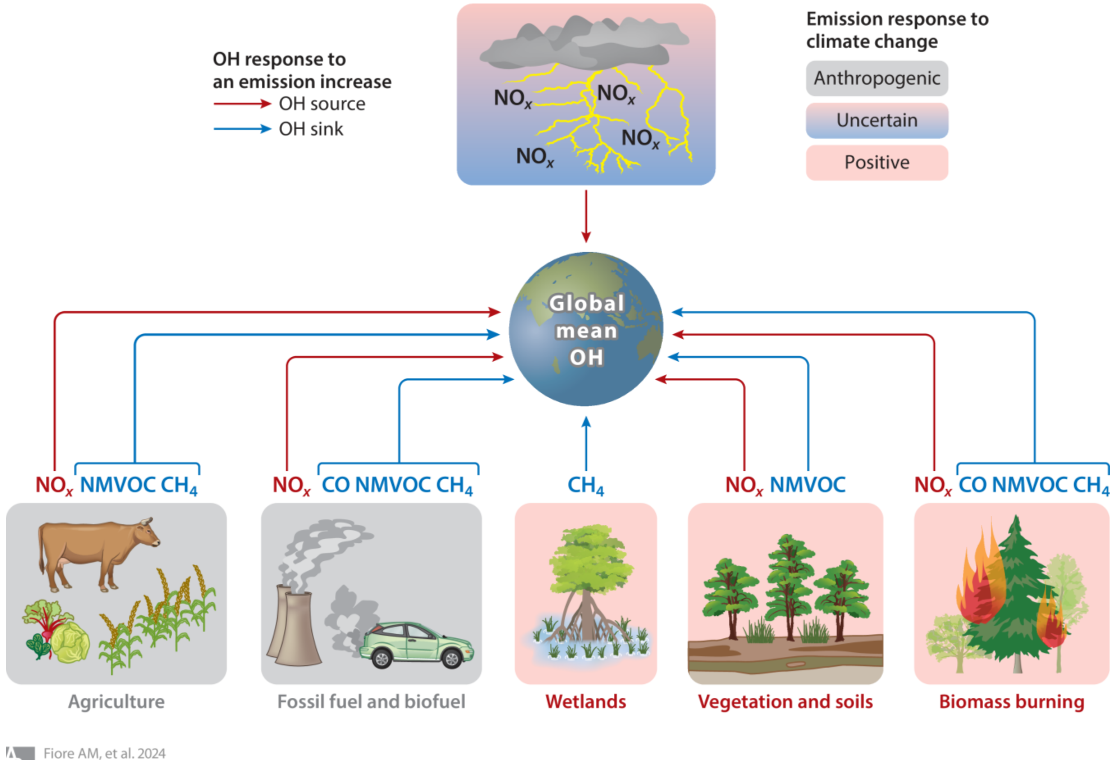


Figure 1: A conceptual illustration of the propagation of uncertainty using concentration and emissions-based anchor points

Sanderson et al., 2024

Reconciliate bottom-up and top-down atmospheric budget estimates (OH)

- **chemistry-climate model** (CCM) **prognostic OH** have uncertainties of around **20 %**
- Uncertainties of **10 % in methyl-chloroform inversions** (e.g., global methane budget 2020).
- CCMs tend to **overestimate the OH distribution**
- Current estimates diverge on the **latitudinal** and **temporal** distribution of OH



Fiore et al., Climate and Tropospheric Oxidizing Capacity, 2024

"mechanistic global atmospheric chemistry transport models fail to even simulate the partitioning of OH between the Northern and Southern Hemispheres ... which alone **warrants further OH studies**."

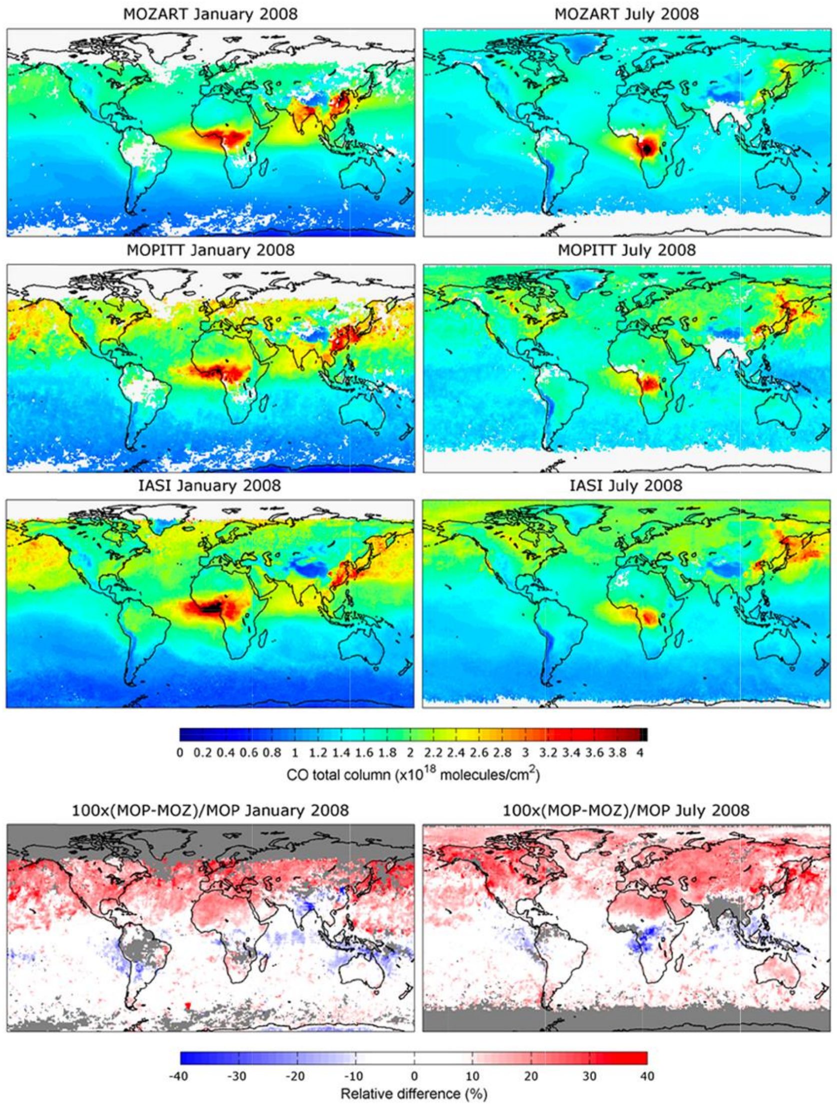
Interpreting contemporary trends in atmospheric methane

Alexander J. Turner[a,1,2], Christian Frankenberg[b,c,1,2], and Eric A. Kort[d,1,2]

Research article | 09 Sep 2014

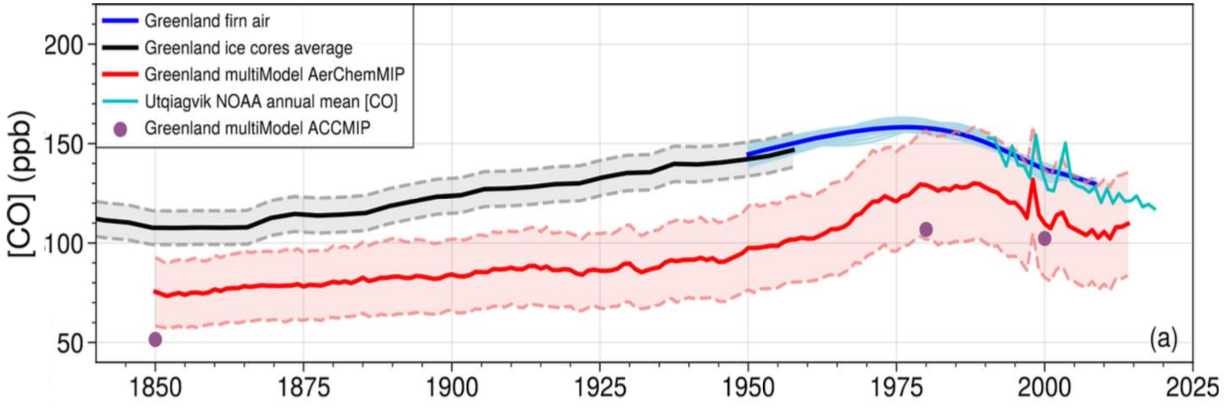
On the wintertime low bias of Northern Hemisphere carbon monoxide found in global model simulations

O. Stein, M. G. Schultz, I. Bouarar, H. Clark, V. Huijnen, A. Gaudel, M. George, and C. Clerbaux



Preindustrial-to-present-day changes in atmospheric carbon monoxide: agreement and gaps between ice archives and global model reconstructions

Xavier Faïn[1], Sophie Szopa[2], Vaishali Naïk[3], Patricia Martinerie[1], David M. Etheridge[4,5], Rachael H. Rhodes[6], Cathy M. Trudinger[4,5], Vasilii V. Petrenko[7], Kévin Fourteau[8], and Philip Place[7,a]



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

Impact of halogen chemistry and methane emissions in CESM2.2 CAM-chem

Community Earth System Model version 2.2 (CESM2.2)
Community Atmosphere Model with chemistry (CAM-chem)

1. **MOZART TS1.2**
2. **MOZART TS1.2 with Short-Lived Halogens (SLH) representation** (Fernandez et al., in prep, 2025)

❖ **CMIP6: Prescribed surface methane concentration** from the CMIP6 protocol

❖ **Methane surface emission flux** from the

GCP2020-Surf: ensemble mean of surface inversions (Saunois et al., 2020)

GCP2020-GOSAT: ensemble mean of GOSAT inversions

CTCH4: CarbonTracker-CH_4-2023 (Oh et al., 2023)

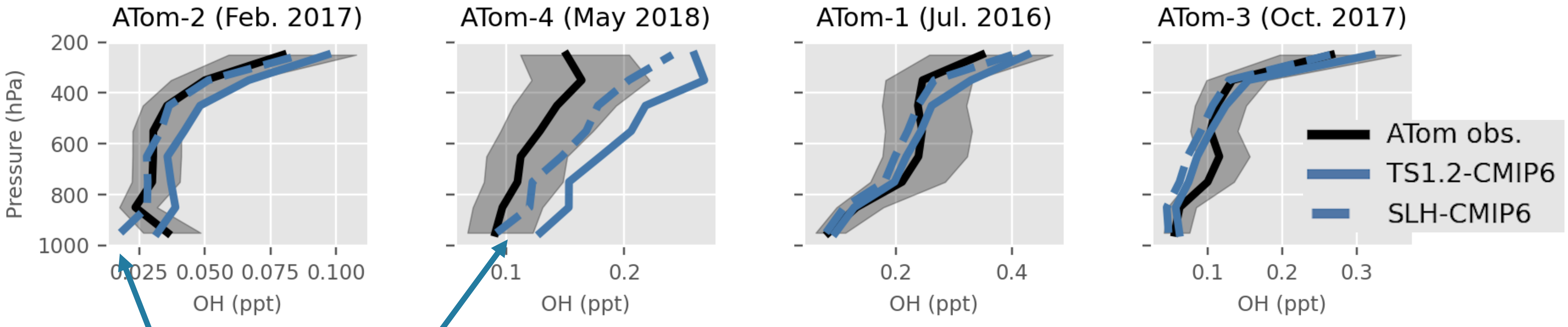
❖ 5-year simulations (January 1st, 2014)

Saunois et al., The Global Methane Budget 2000–2017, Earth Syst. Sci. Data, 2020

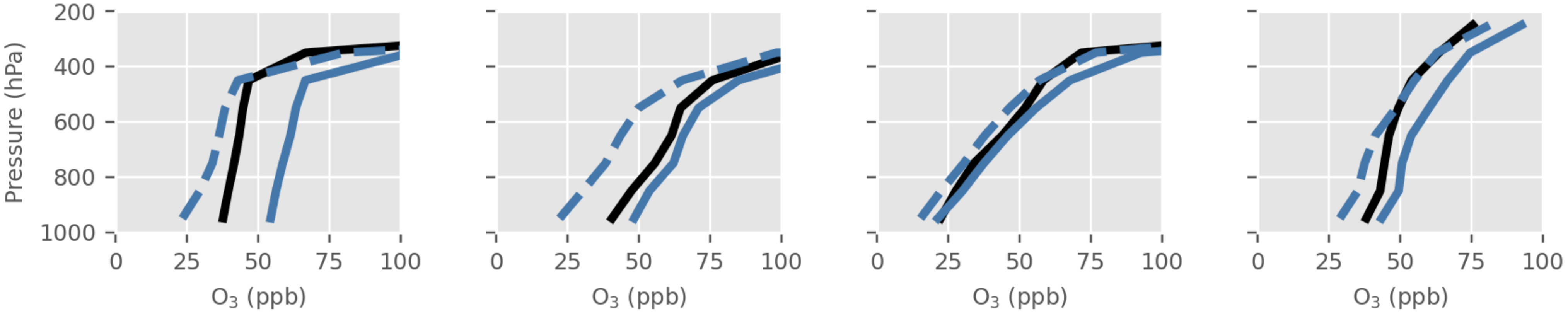
Oh, Y., et al.,. CarbonTracker CH_4 2023. NOAA Global Monitoring Laboratory. https://doi.org/10.25925/40JT-QD67, 2023

Impact of halogen chemistry and methane emissions in CESM2.2 CAM-chem

NASA ATom Latitudes > 30°N

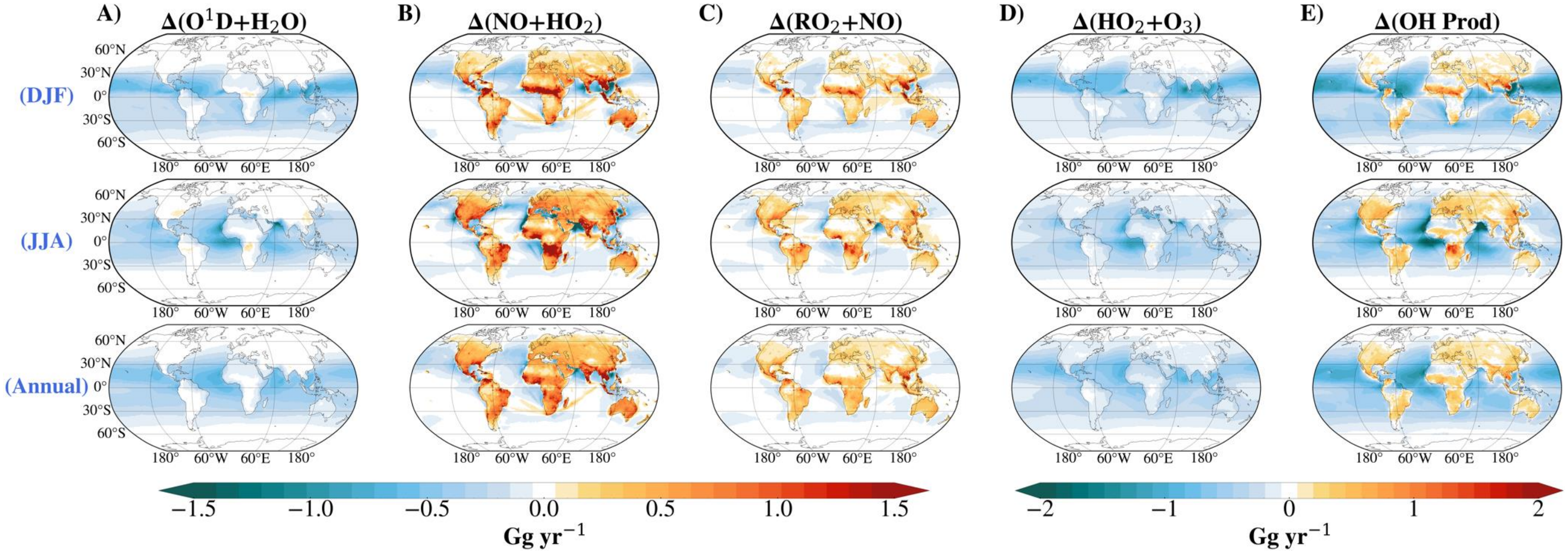


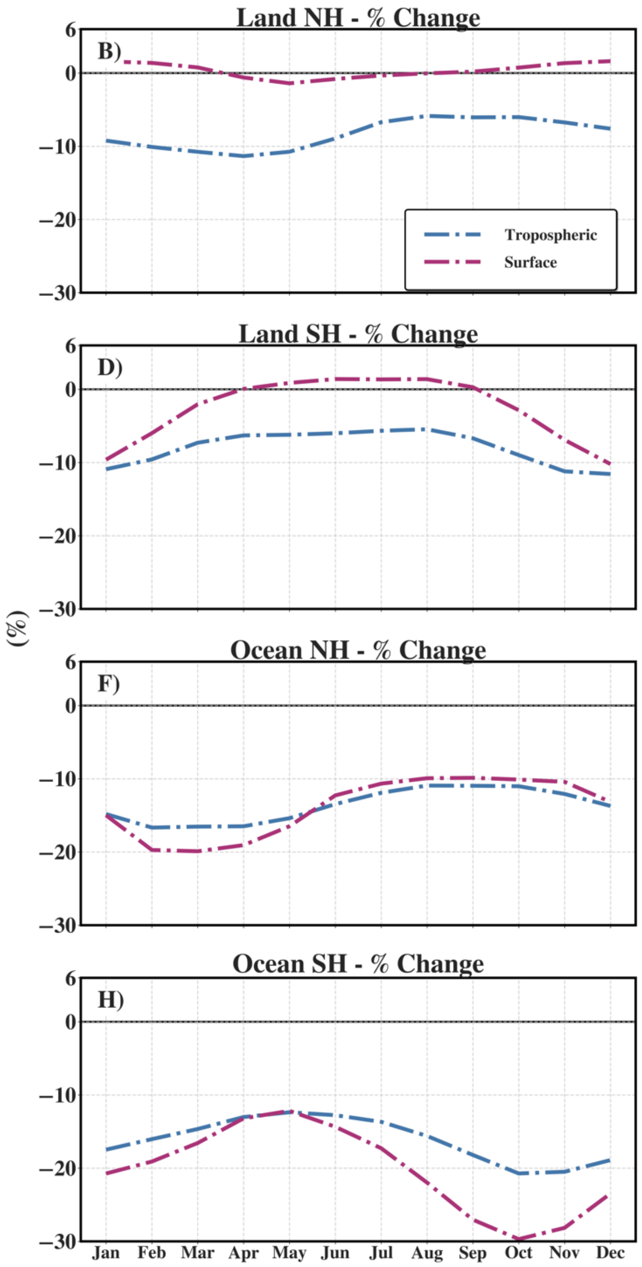
Improved OH in winter and spring



O_3 reduction in winter and spring

Impact of halogen chemistry and methane emissions in CESM2.2 CAM-chem





Over land:

OH does not changes at the surface

tropospheric OH decreases by up to 10 %

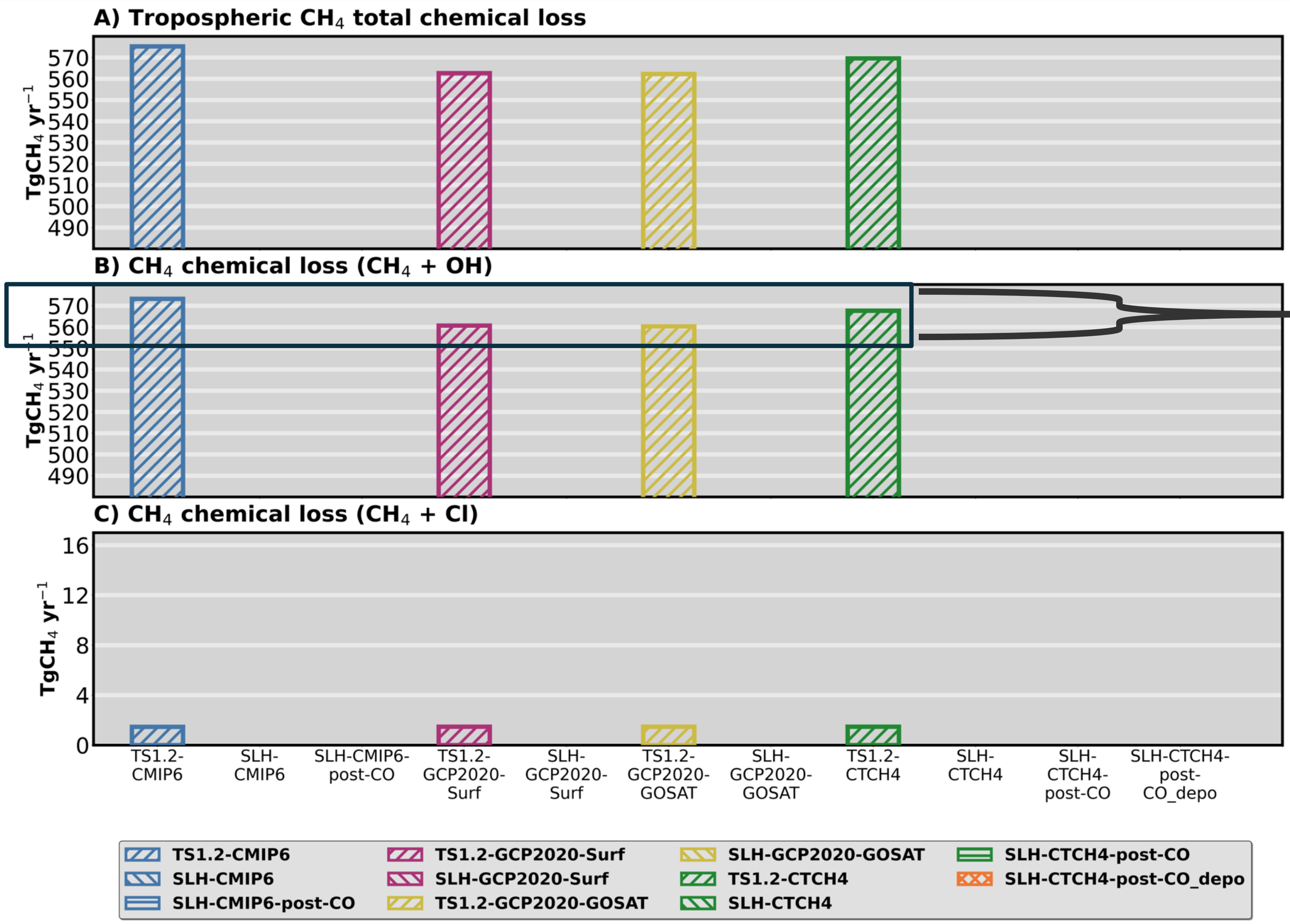
Over the ocean:

OH is reduced by up to 20 % at the surface

tropospheric OH decreases range between 10 % and 20 %

A. Mirrezaei et al, to be submitted, 2025

Methane Chemical loss budget (2017)



MOZART TS1.2

Default chemistry MOZART TS1.2

TS1.2-CMIP6: 573 $TgCH_4\ yr^{-1}$

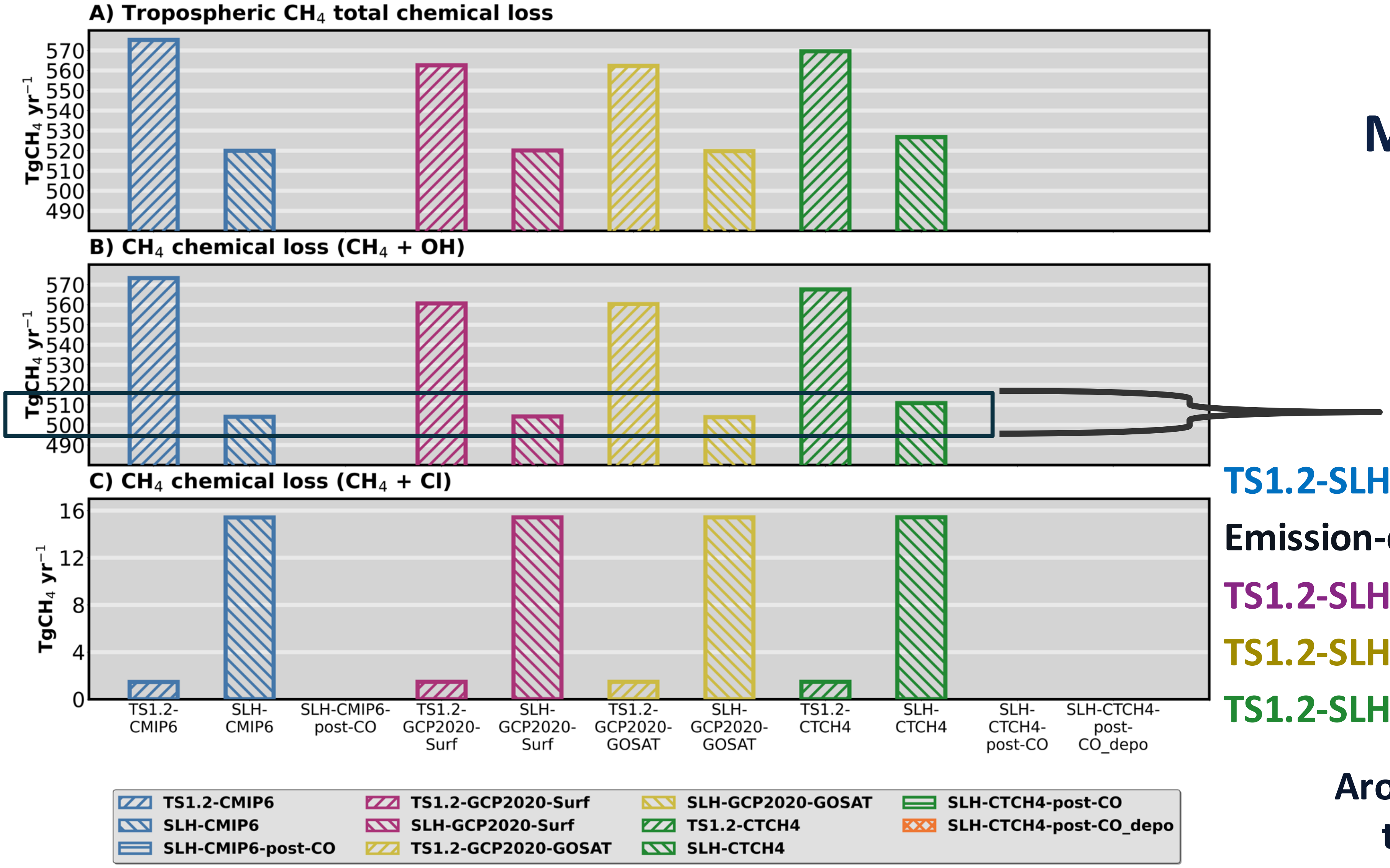
Emission-driven simulations:

TS1.2-GCP2020-Surf: 561 $TgCH_4\ yr^{-1}$

TS1.2-GCP2020-GOSAT: 561 $TgCH_4\ yr^{-1}$

TS1.2-CTCH4: 568 $TgCH_4\ yr^{-1}$

Methane Chemical loss budget (2017)



MOZART TS1.2 SLH

CH_4+OH TS1.2-SLH

TS1.2-SLH-CMIP6: 504 $TgCH_4\ yr^{-1}$

Emission-driven simulations:

TS1.2-SLH-GCP2020-Surf: 504 $TgCH_4\ yr^{-1}$

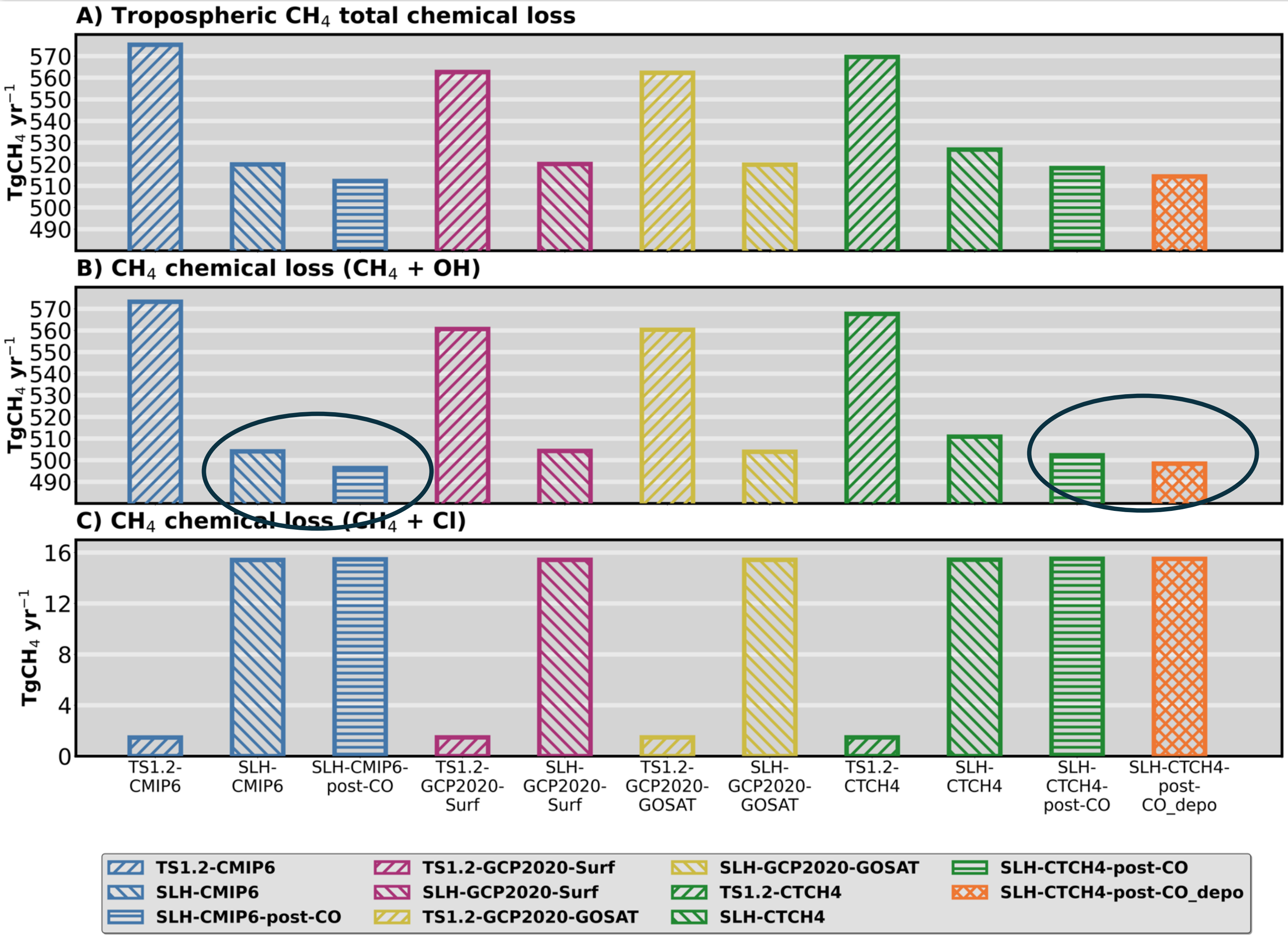
TS1.2-SLH-GCP2020-GOSAT: 504 $TgCH_4\ yr^{-1}$

TS1.2-SLH-CTCH4: 511 $TgCH_4\ yr^{-1}$

Around 16 $TgCH_4\ yr^{-1}$ from the tropospheric CH_4+Cl sink

The CH_4 tropospheric chemical lifetime (OH) increase from 8.82-8.88 to 10.11-10.14 years

Methane Chemical loss budget (2017)



Impact of CO emissions (net increase by 180 TgCO yr^{-1}

TS1.2-SLH-CMIP6: 504 $TgCH_4\ yr^{-1}$

TS1.2-SLH-CMIP6-post-CO: 496 $TgCH_4\ yr^{-1}$

TS1.2-SLH-CTCH4: 511 $TgCH_4\ yr^{-1}$

TS1.2-SLH-CTCH4-post-CO: 502 $TgCH_4\ yr^{-1}$

Around 8 to 9 $TgCH_4\ yr^{-1}$ from a change in CO emissions

Revisiting an old conundrum: Tropospheric Ozone burden vs OH

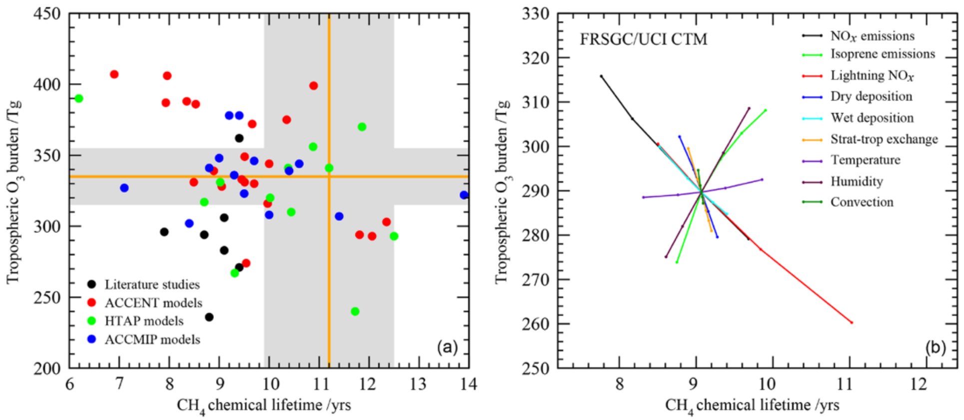
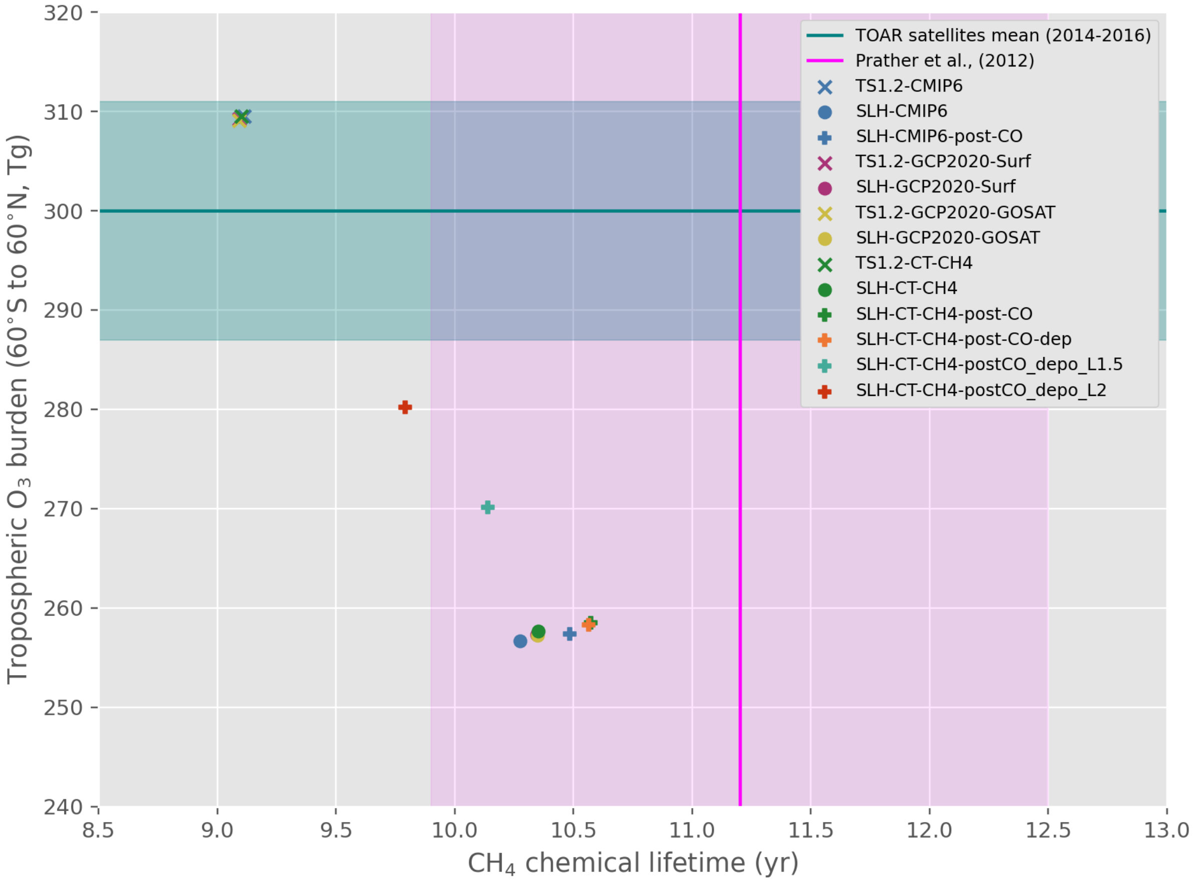


Figure 1. Tropospheric oxidant budgets from previous published studies and model intercomparisons **(a)**, along with measurement-based estimates of the tropospheric O_3 burden and CH_4 lifetime (shaded regions). Panel **(b)** shows results from one-at-a-time sensitivity studies with a single model revealing 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

Impact of methane emissions in CESM2.2 CAM-chem

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Community Atmosphere Model with chemistry (CAM-chem)

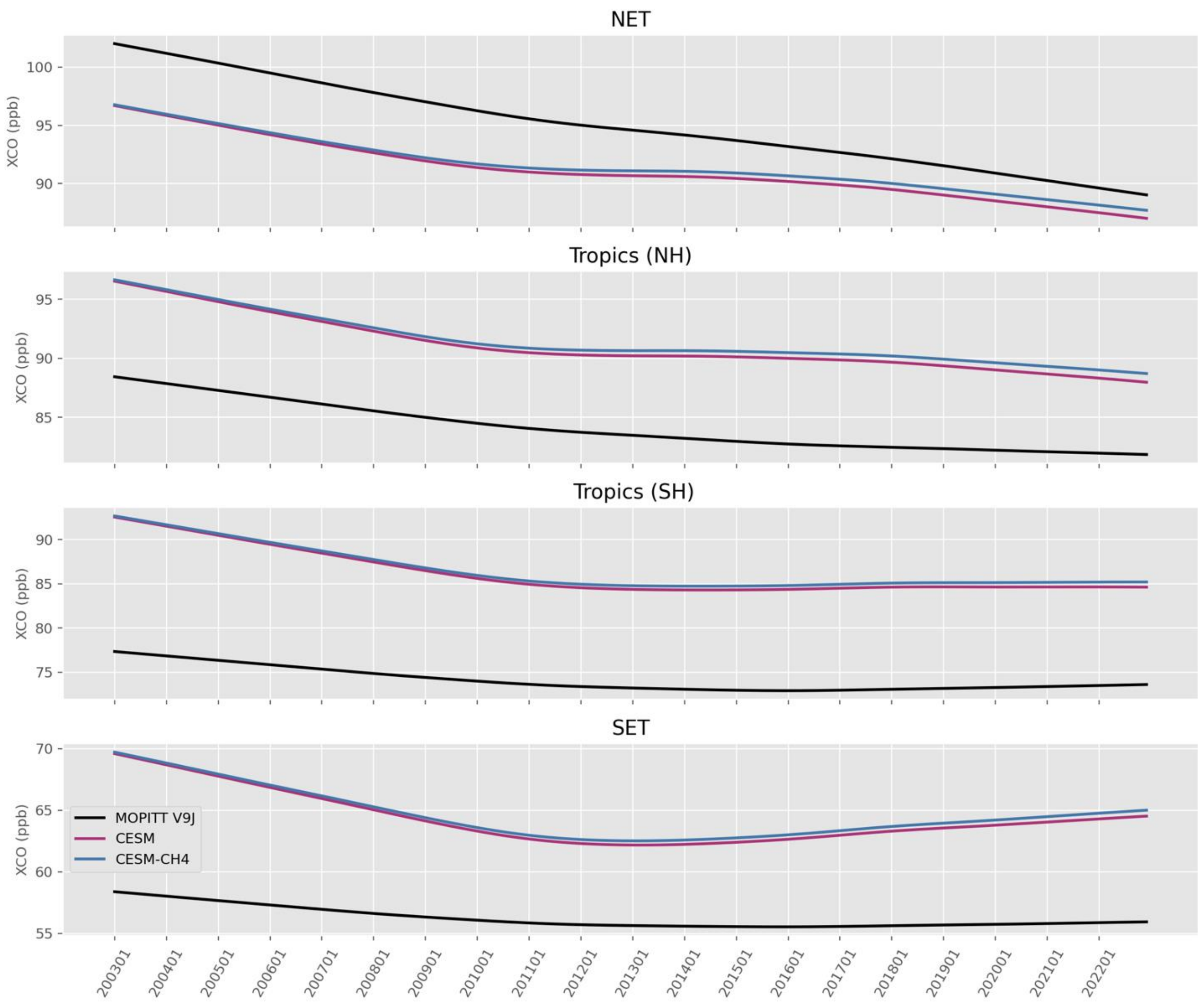
MOZART TS1.2 with improved Short-Lived Halogens (SLH) representation (Fernandez et al., in prep, 2025)

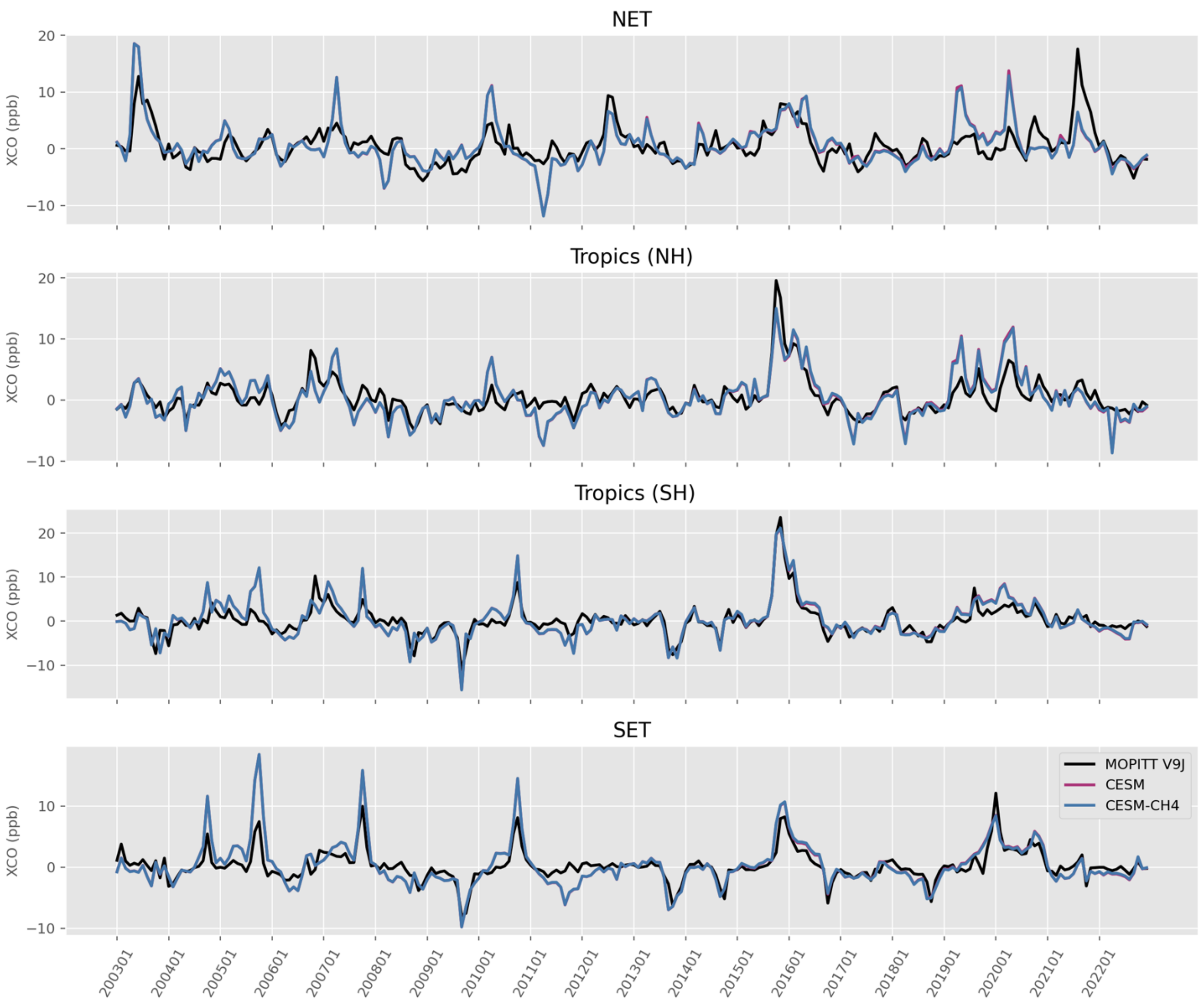
- ❖ **CESM: Prescribed surface methane concentration** from the CMIP6 protocol
- ❖ **CESM-CH4: Methane surface emission flux** from the CarbonTracker-CH_4-2025 (Oh et al., 2023)

- ❖ 20-year simulations (2003 to 2022)

Oh, Y., et al.,. CarbonTracker CH_4 2025. NOAA Global Monitoring Laboratory. https://doi.org/10.25925/40JT-QD67, 2023

MOPITT CO: Seasonal and Trend decomposition using Loess (STL)

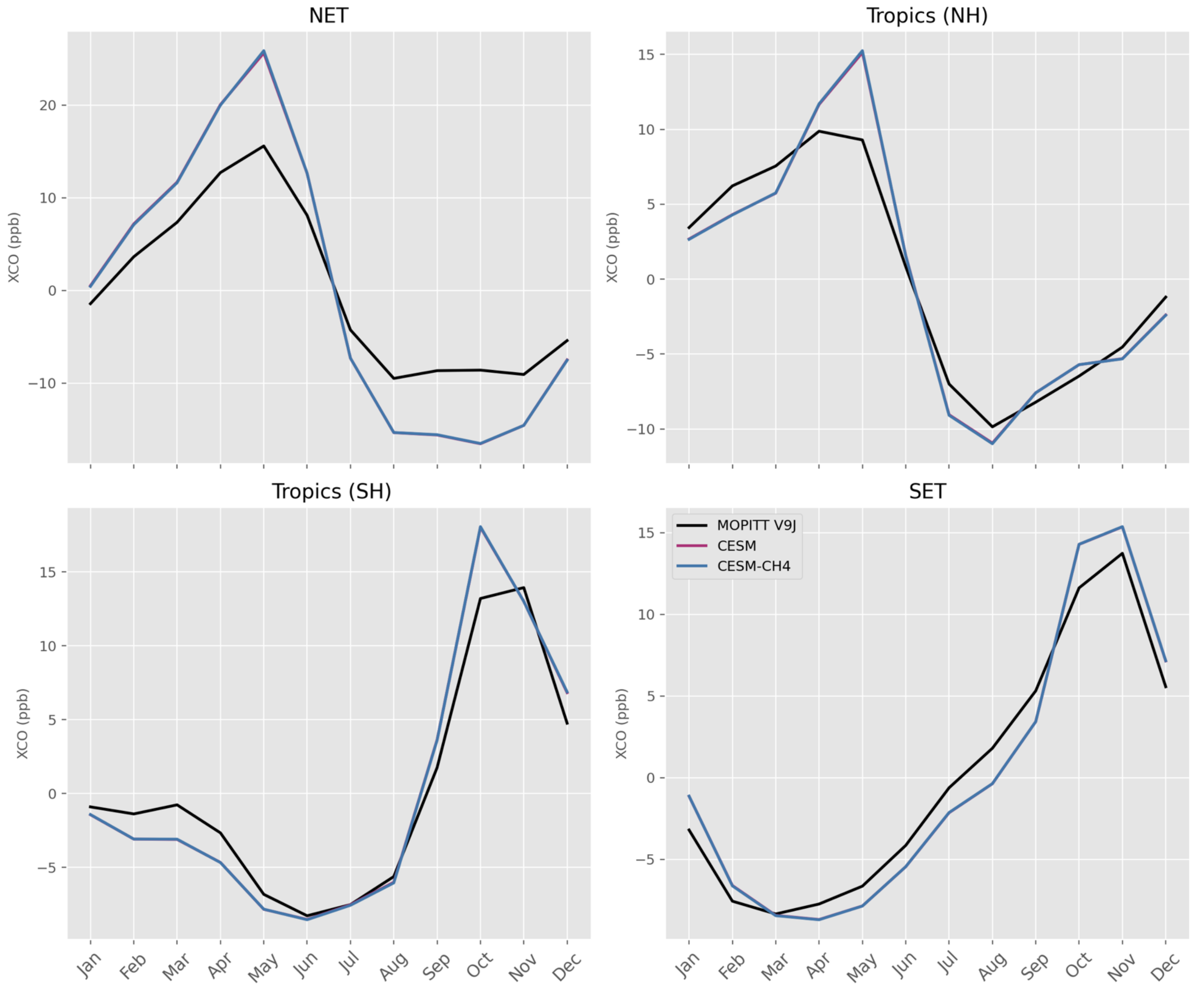




Trend component

Inter-annual variability component

MOPITT CO: Seasonal and Trend decomposition using Loess (STL)

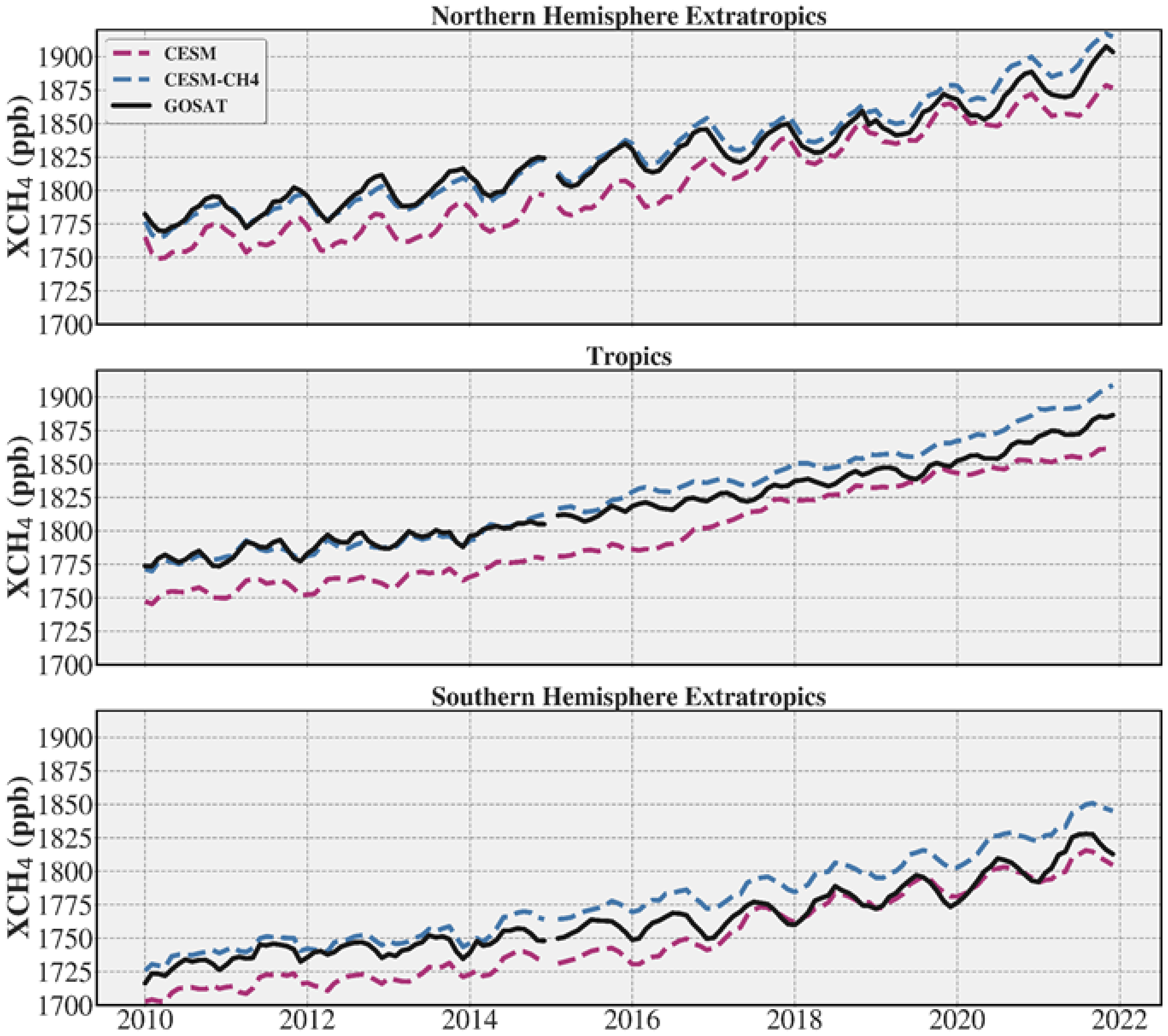


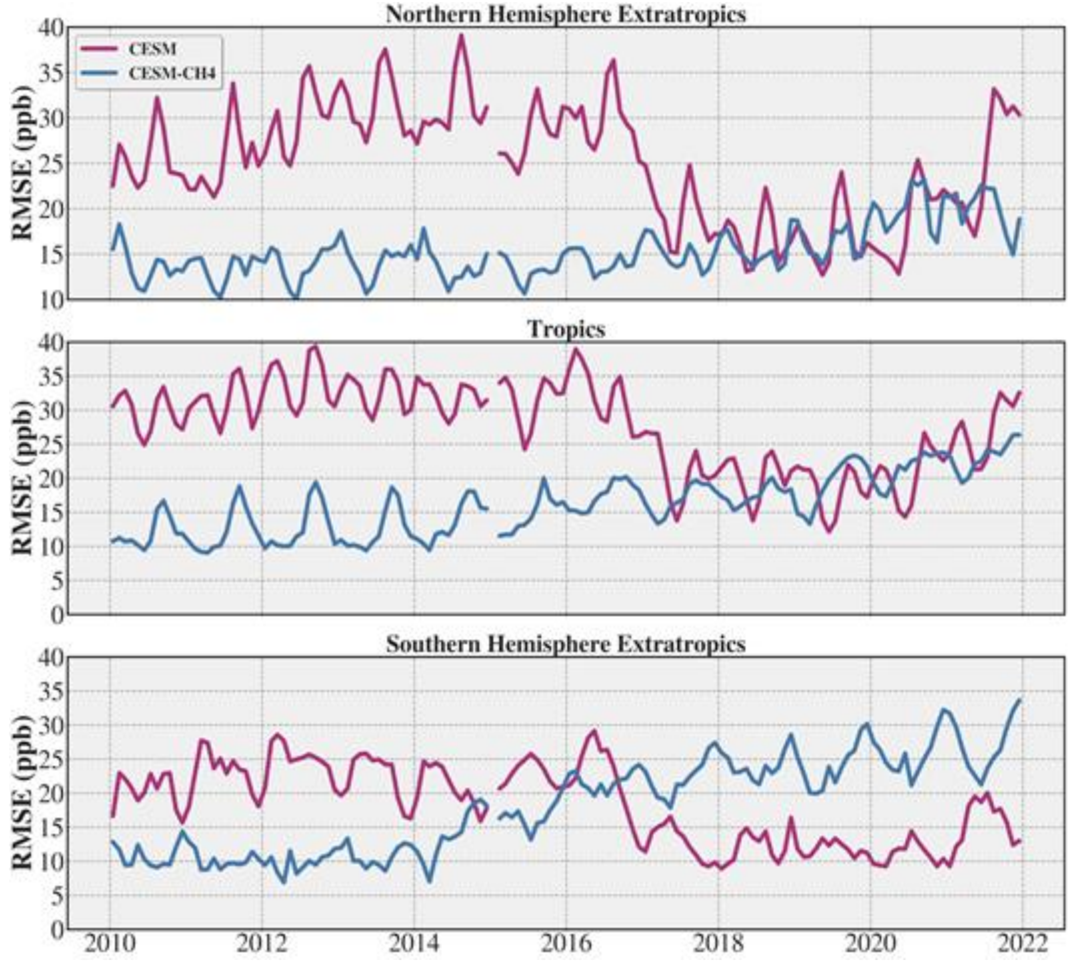
Average Seasonal Cycle (2003-2022)

Summertime underestimation and spring overestimation

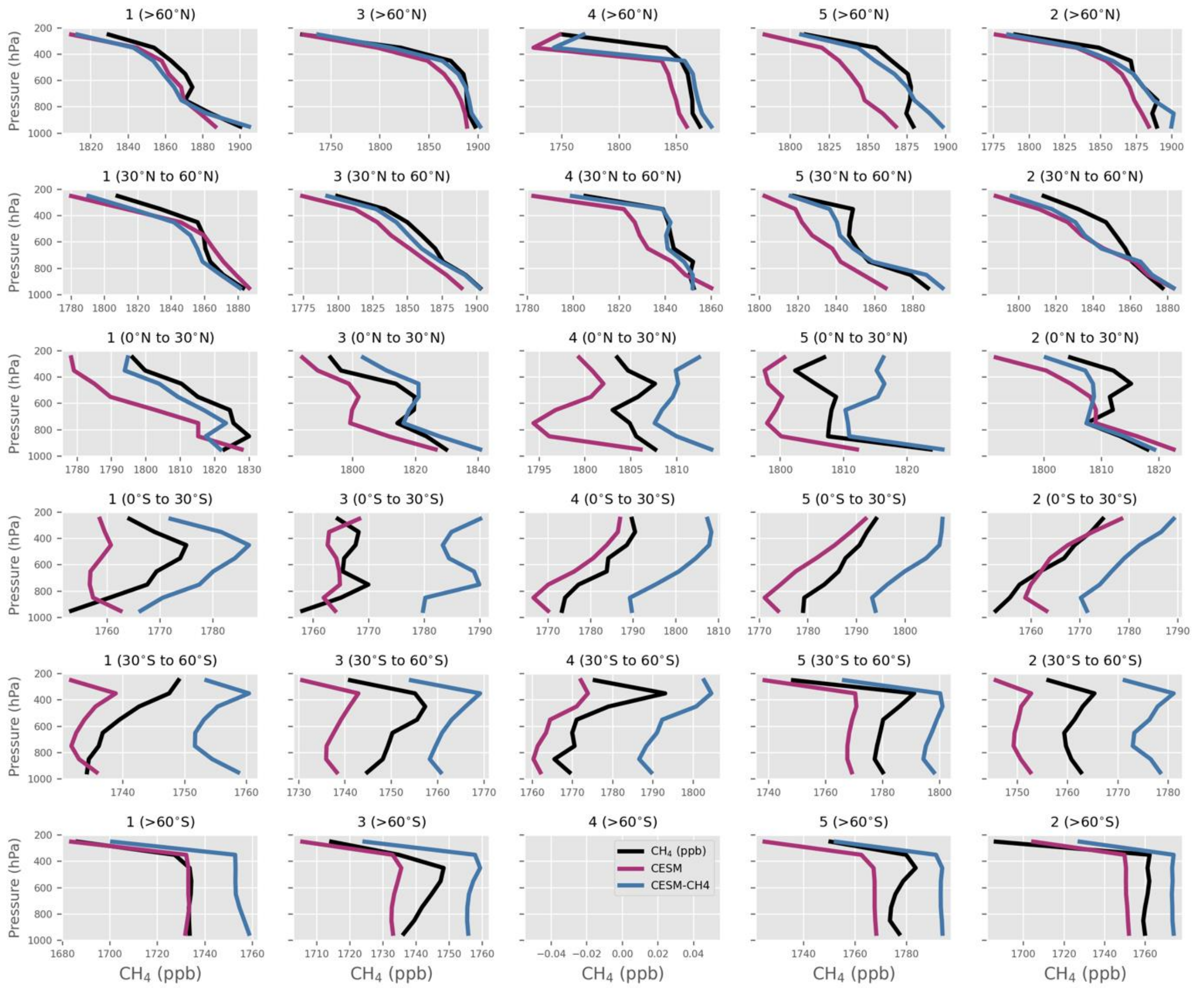
- Photochemistry (CO+OH loss)
- Secondary CO source, i.e., Volatile Organic Compounds chemistry
- Summertime emissions from fires, biogenic and anthropogenic sources.

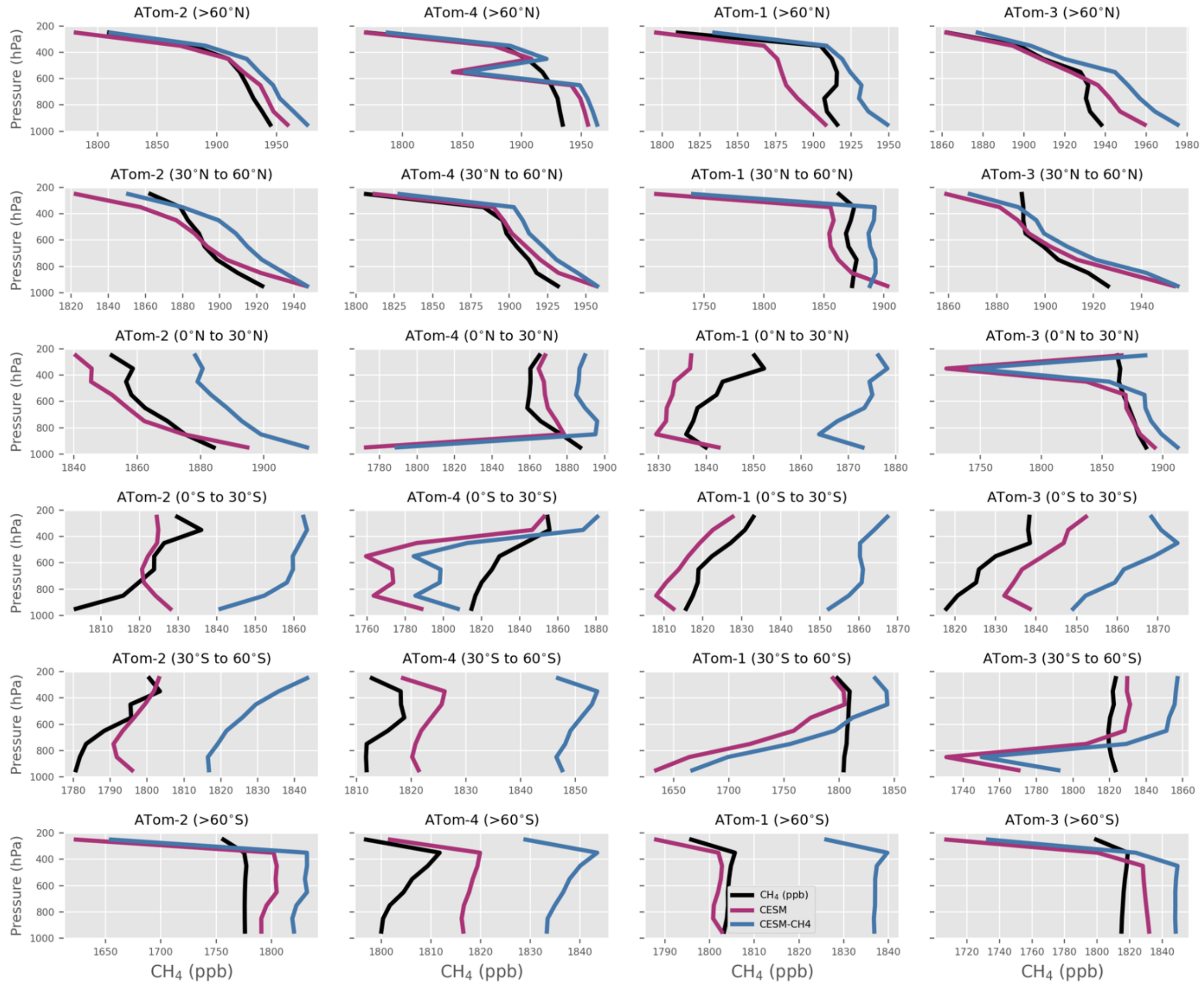
GOSAT CH_4 (2010 to 2022)





NSF HIPPO (2009-2011) and NASA ATom (2016-2018)

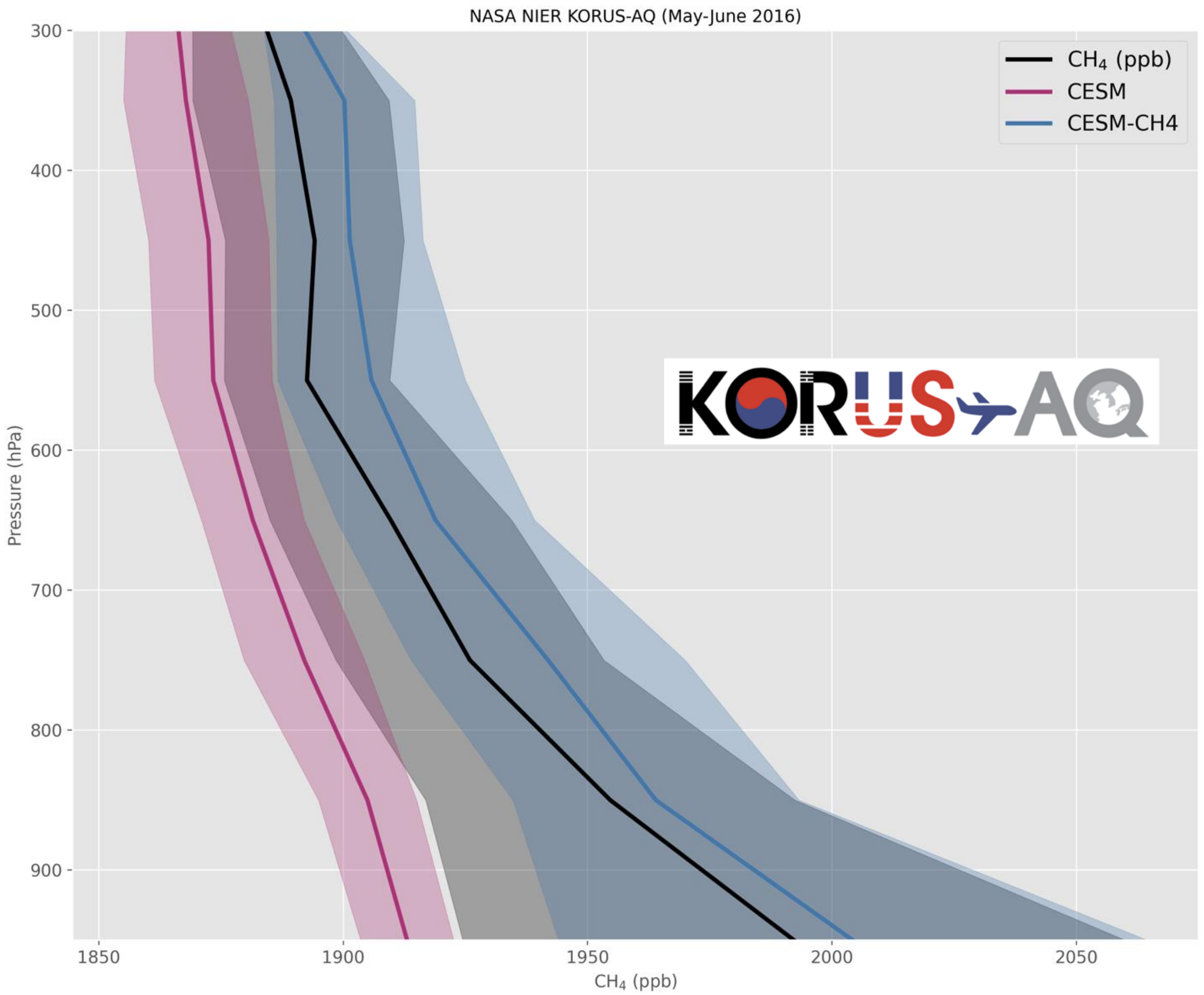


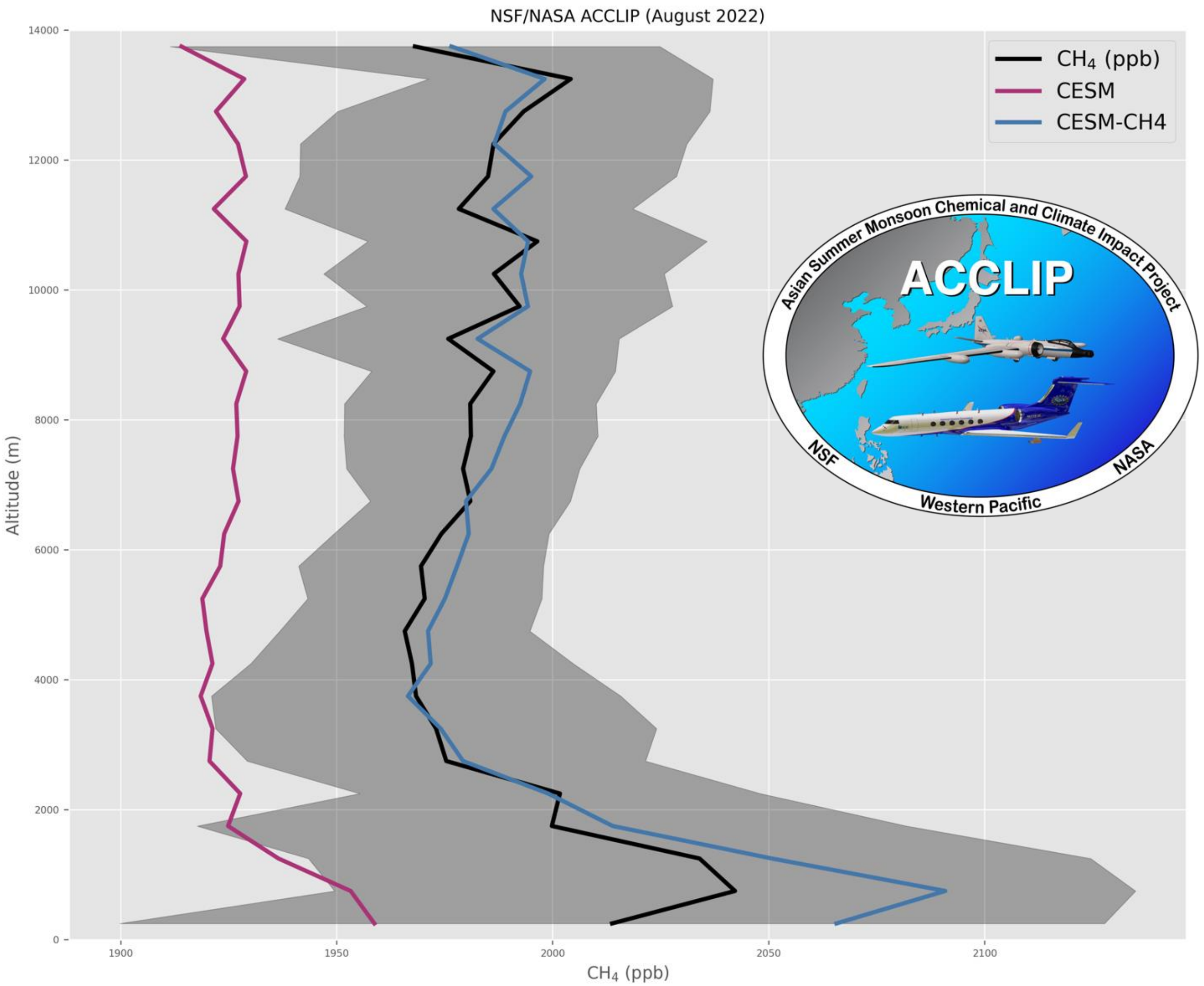


Five campaigns 2009-2011, transecting Pacific

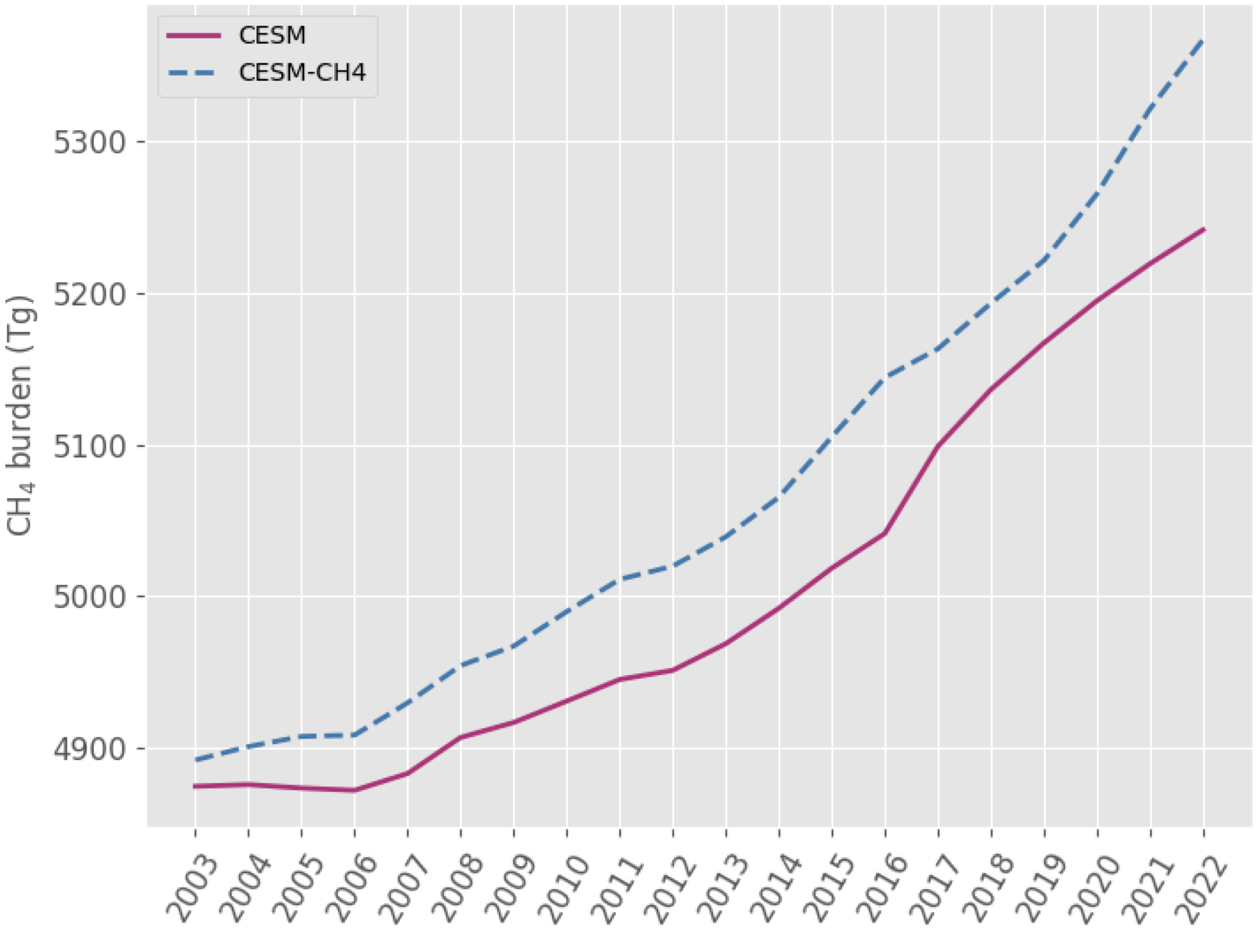
Four campaigns 2016-2018, transecting Pacific and Atlantic

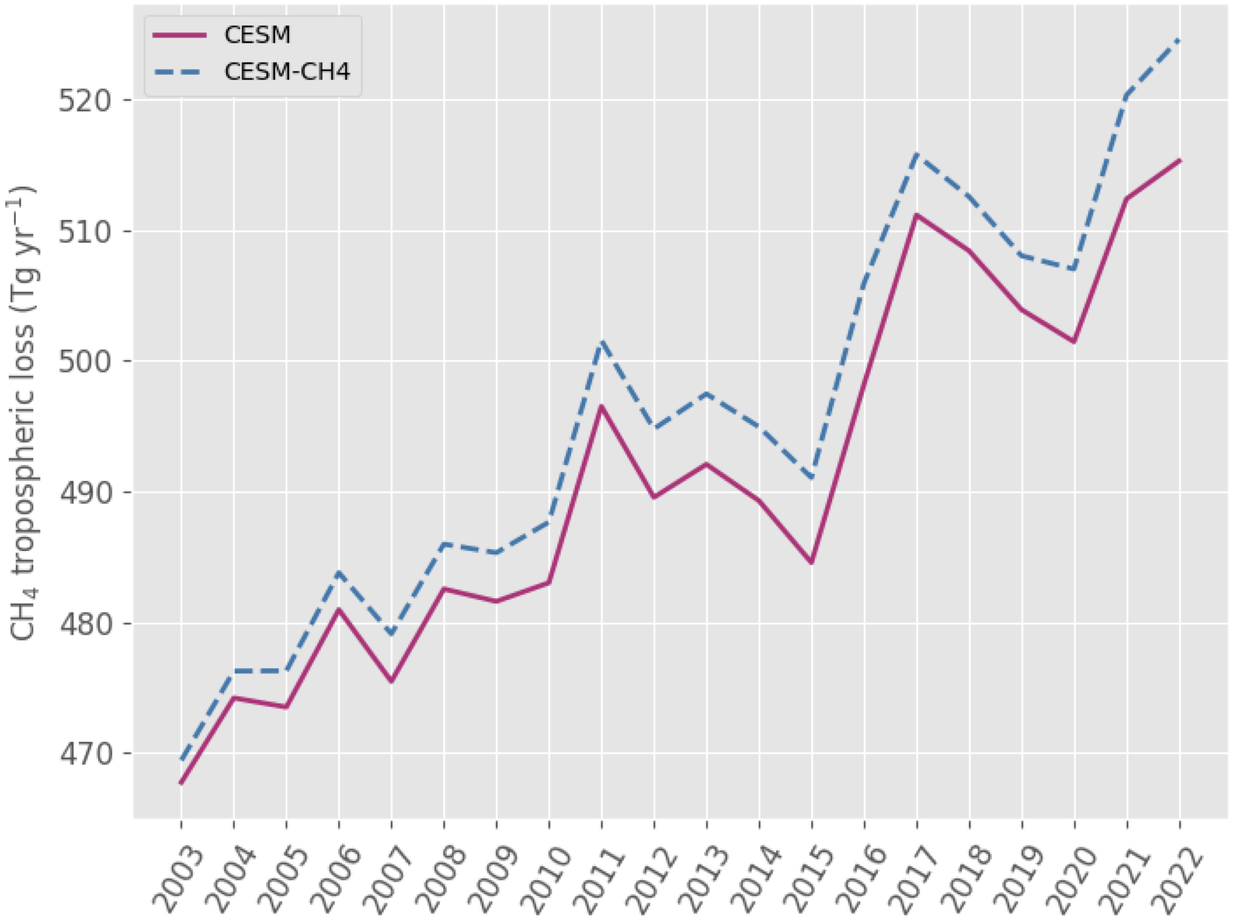
KORUS-AQ (2016) and ACCLIP (2022)





Methane Budget





➢ CH_4 chemical loss is well buffered in our improved chemistry

Summary

- Updated halogen representation in CESM2.2 impacts:
  - ✓ Net reduction in O_3, OH with most profound impact over the ocean
  - ✓ Result in an increase in CO and CH_4 due to a longer lifetime with respect to OH
- Dramatic improvements in CO and CH4 simulations
  - ✓ Evaluated against GOSAT XCH_4, MOPITT XCO, airborne field campaign observations
  - ✓ Latitudinal gradient in biases, southern hemisphere CO/CH_4 overestimation

- Enables emission-driven 20 year simulation transient simulations
- CH_4 chemical loss is well buffered in our improved chemistry

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