Investigating the impact of uncertainties in CH_4 emissions and halogen chemistry on CH_4 abundance and lifetime

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Background

Methane Emissions in Chemistry-Climate Models

- Methane concentration in atmosphere is a complex balance of sources and sinks.
- Chemistry-Climate Models commonly employ a global time-varying methane surface concentration (LBC) pre-calculated from emissions.
- LBC methane buffers atmospheric oxidation's effect on methane levels at the surface
- An interactive chemistry approach is needed to capture methane's interactions with oxidants like OH
- Few studies use methane emissions in Chemistry-Climate Models

Heimann, I., et al. "Methane emissions in a chemistry-climate model: Feedbacks and climate response." Journal of Advances in Modeling Earth Systems 12.10 (2020)

Background

Updated Short-Lived Halogens (SLH) representation

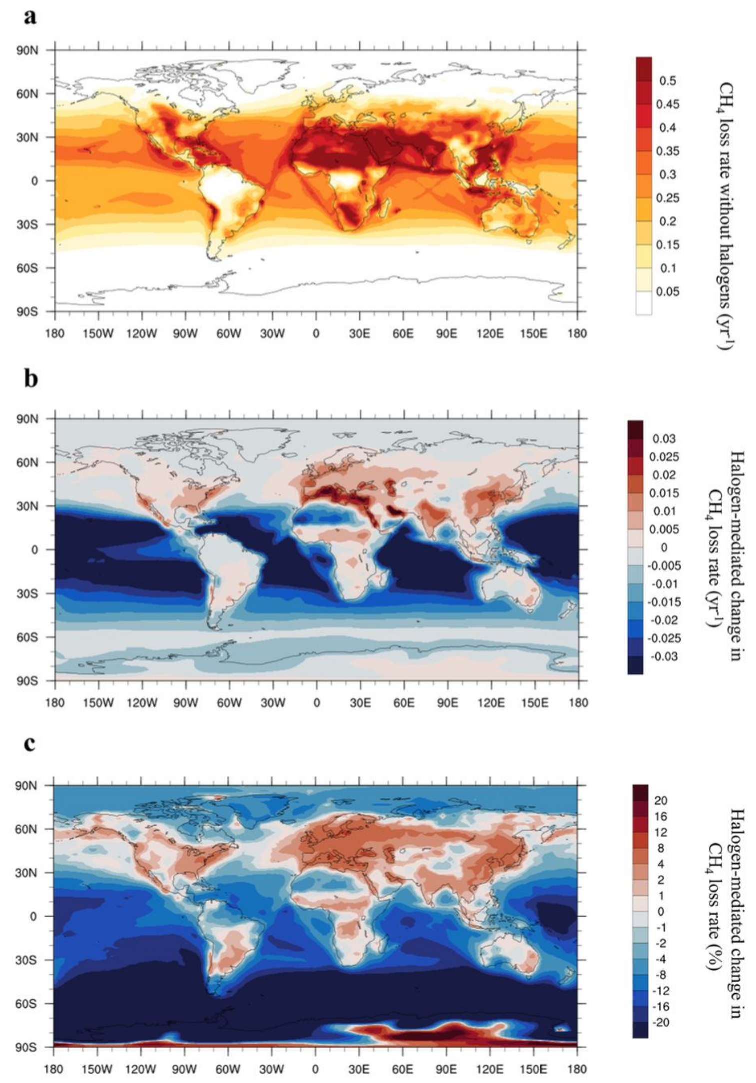
- Halogenated species affect tropospheric oxidative capacity and ozone and hydroxyl radical (OH) budgets.

- Methane oxidation:
  $CH_4 + Cl \rightarrow CH_3O_2 + HCl$
  $CH_4 + OH \rightarrow CH_3O_2 + H_2O$

- VSL Halogens indirectly decrease OH by destroying O_3, the main source of the OH

- The reduction of CH_4 loss, increases the lifetime of CH_4 in the atmosphere.

Li et al., Reactive halogens increase the global methane lifetime and radiative forcing in the 21st century, nature communication, 2022



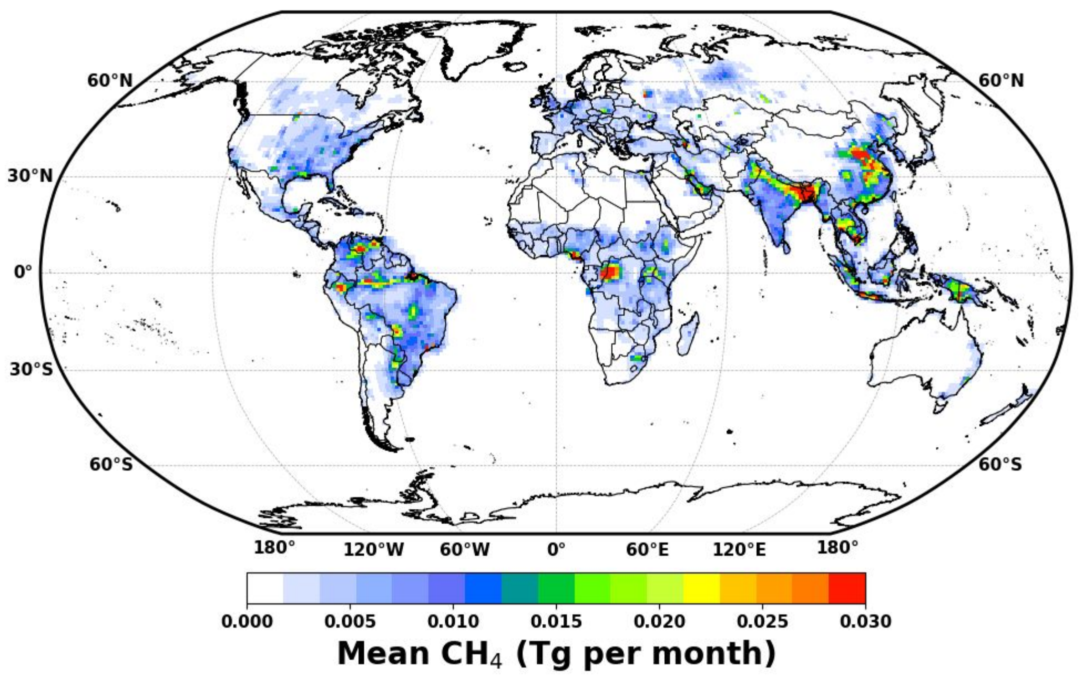
Impact of Halogen Chemistry and Methane Emissions in CESM2.2 CAM-chem

- Number of Simulations=7
- Methane emission from the Global Carbon Project 2020 (GCP2020, Saunois et al., 2020), instead of prescribed surface concentration from the CMIP6 protocol.
- Very Short Lived (VSL) halogen emissions and chemistry, applied to CESM2.2

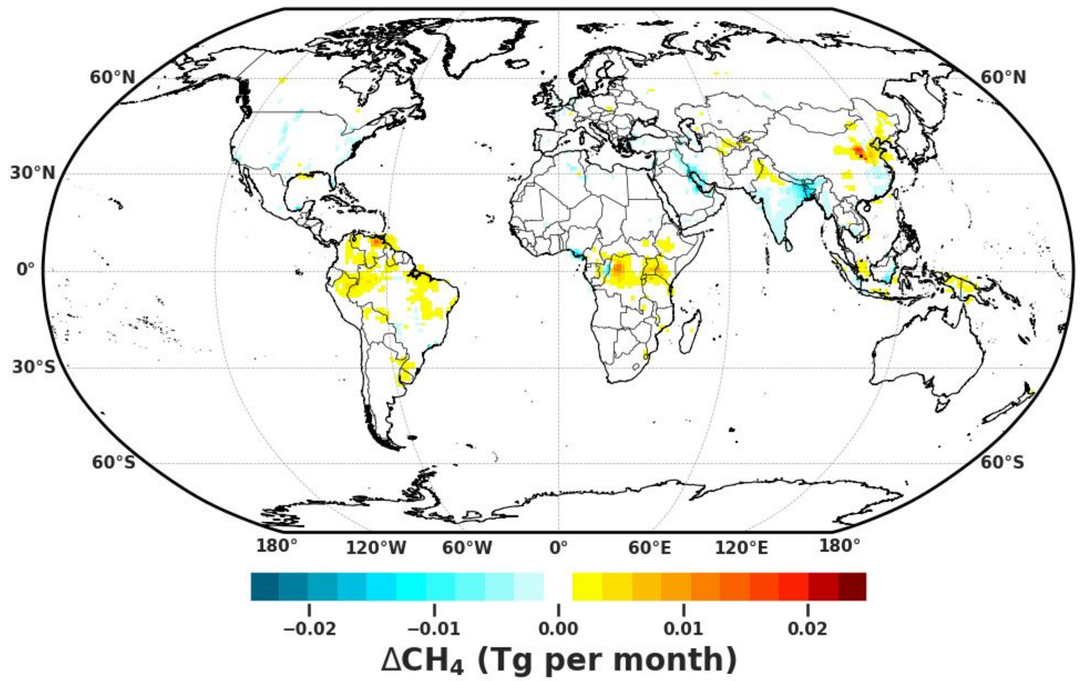
Simulations	Chemistry	Methane	Anthro CO emission	Fire CO emission
TS1	TS1	prescribed (CMIP6)	CAMS-GLOB-ANT_v5.3	FINN2.5
TS1-GCP-2020-SURF/ GOSAT	TS1	GCP2020-SURF	CAMS-GLOB-ANT_v5.3	FINN2.5
TS1-GCP-2020-SURF/ GOSAT	TS1	GCP2020-SURF	CAMS-GLOB-ANT_v5.3	FINN2.5
TS1-VSL	**TS1-VSL**	prescribed (CMIP6)	CAMS-GLOB-ANT_v5.3	FINN2.5
TS1-VSL GCP-2020-SURF/GOSAT	**TS1-VSL**	GCP2020-SURF	CAMS-GLOB-ANT_v5.3	FINN2.5
TS1-VSL GCP-2020-SURF	**TS1-VSL**	GCP2020-SURF	MOPITT inversion	FINN2.5

GOSAT and Surface GCP 2020 inversion CH_4 emissions

Mean GOSAT and Surface CH_4 Emissions

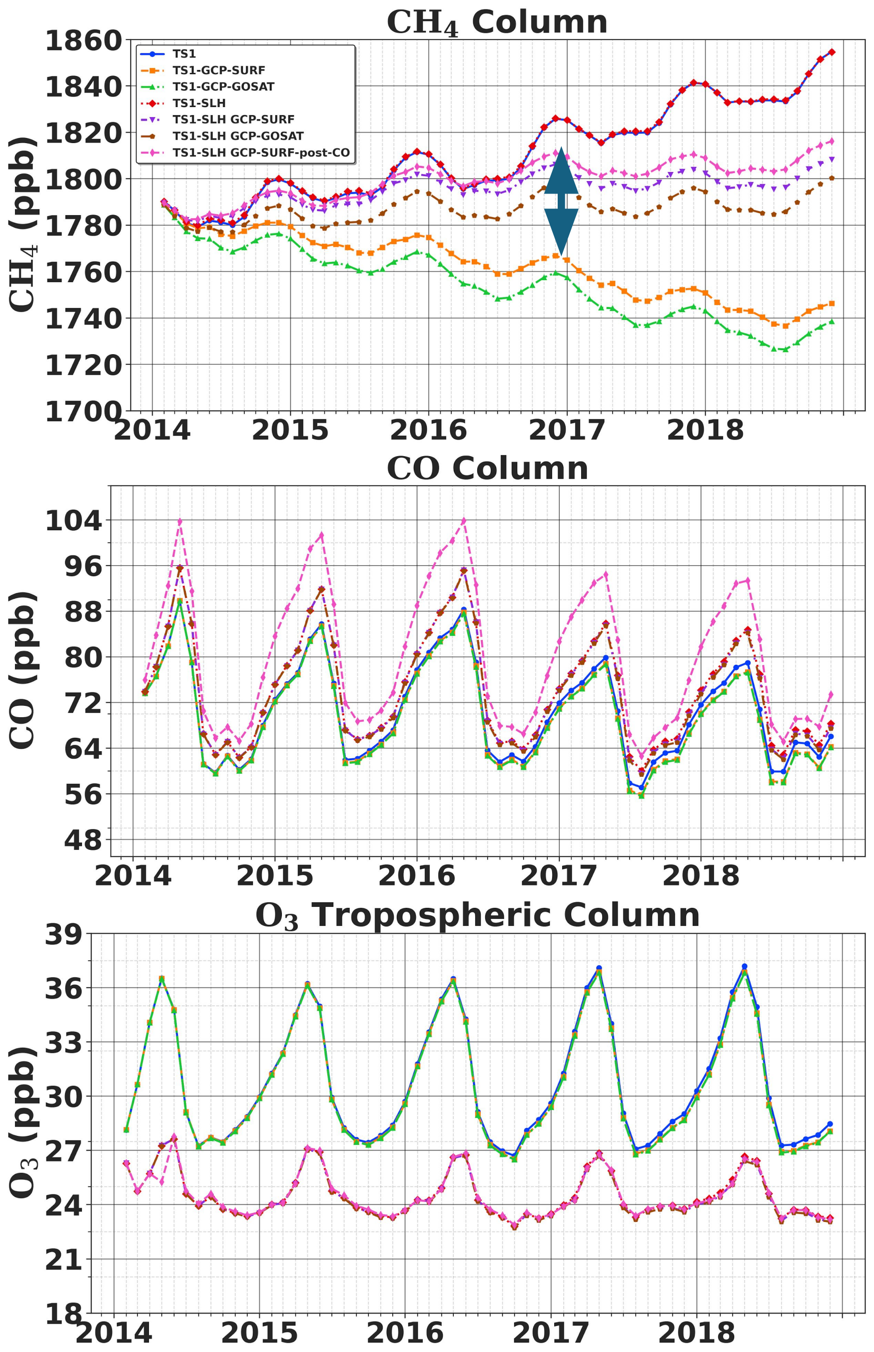


GOSAT- Surface Emissions



Zonal averages of temporal change of CH_4, CO, and O_3

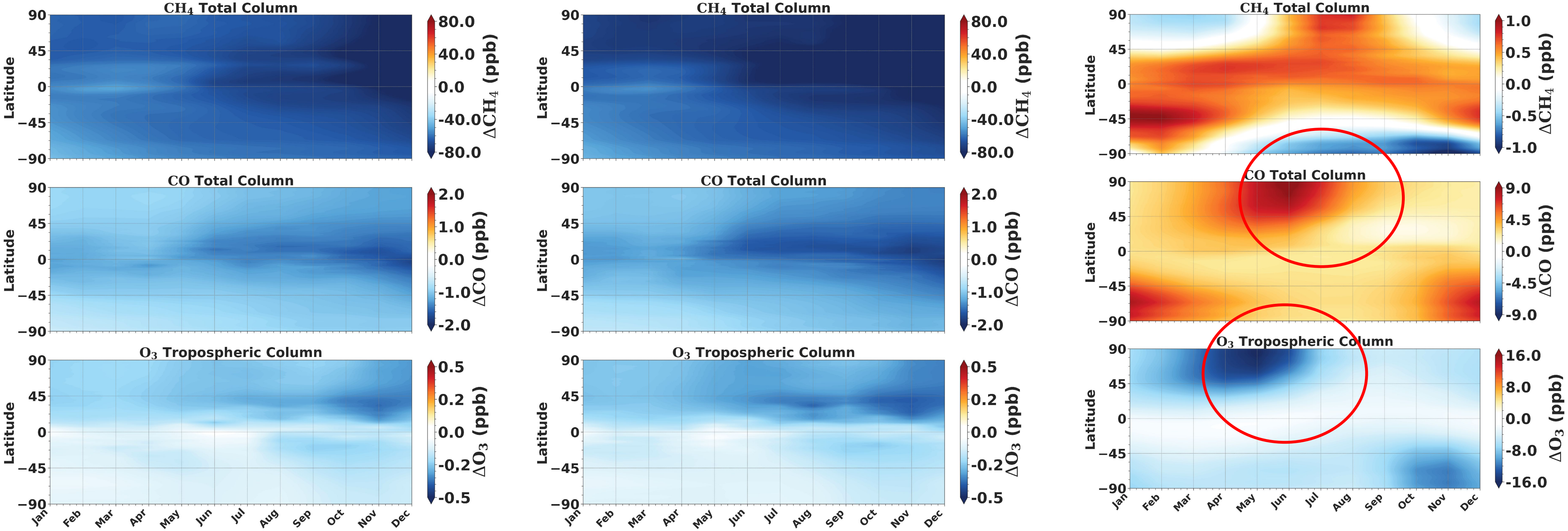
- Emission-driven simulations without updated halogen chemistry show significant decrease in CH_4 total column compared to simulations with prescribed CH_4
- Use of posterior CO emissions improves CH_4
- Emission-driven methane simulations with updated halogen chemistry improves CO



Relative change of CH_4, CO, and O_3 compared to TS1

- CH_4 emission driven simulations show a **significant decrease** in CH_4 total column
- CH_4 emission driven simulations does not have significant impact on **CO** and **O_3**
- Simulation with revised halogen chemistry shows an increase in CH_4 and CO columns and a decrease in the O_3 tropospheric column.

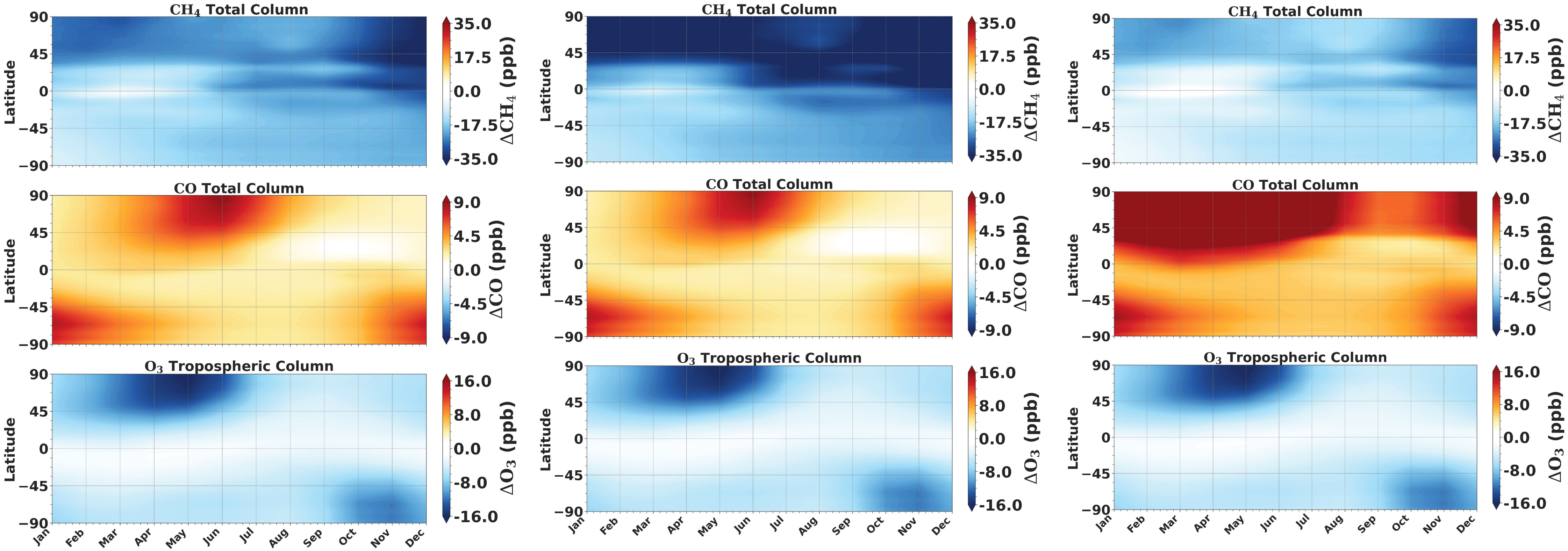
TS1-GCP-SURF - TS1	TS1-GCP-GOSAT - TS1	TS1-SLH - TS1



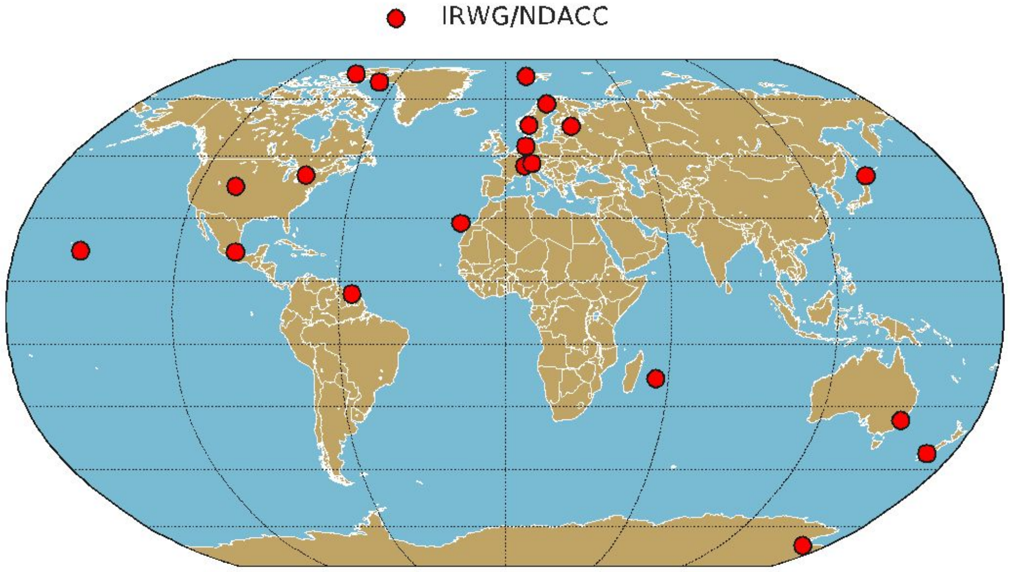
Relative change of CH_4, CO, and O_3 compared to TS1

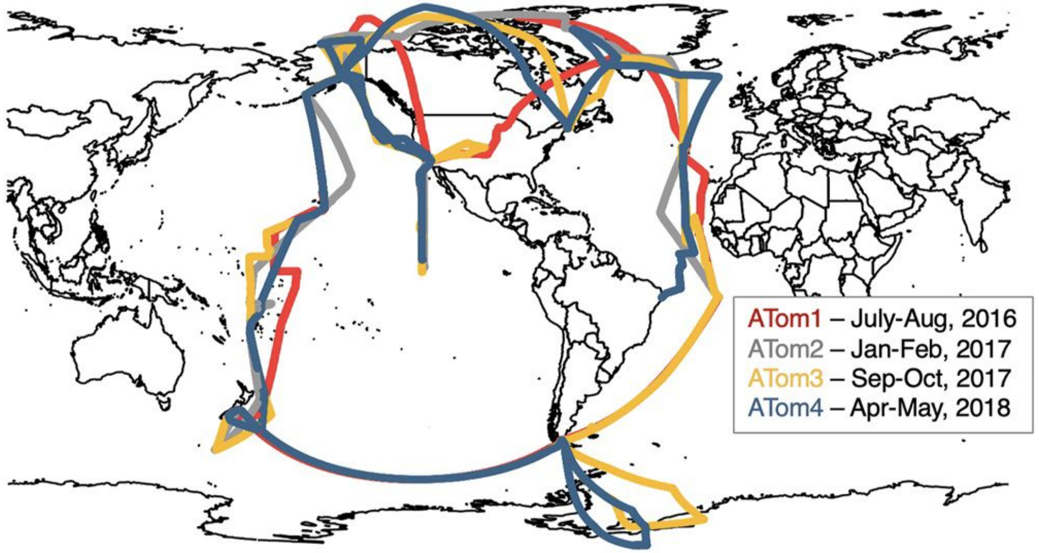
- Using updated halogen chemistry **reduces drop** in **CH_4** total column in the emission driven simulations
- Using posterior CO emissions and improved halogen chemistry improve CH_4 total column
- Simulation with updated halogen chemistry shows **sensitivity** to **CH_4 emission**

TS1-GCP-SURF - TS1	TS1-GCP-GOSAT - TS1	TS1-SLH - TS1



NASA Atmospheric Tomography Mission (ATom) and NDACC sites





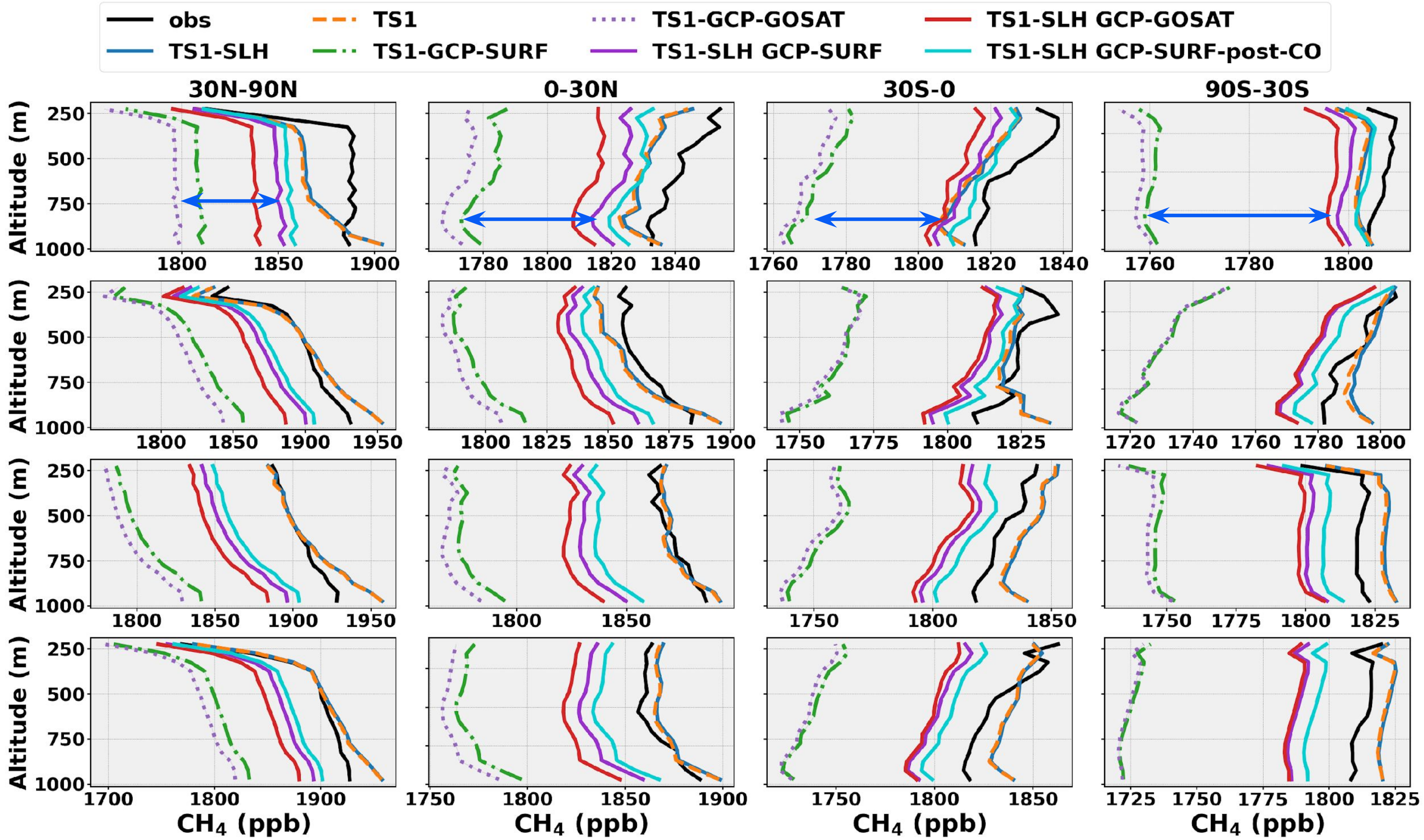
CH_4 and CO

CH_4 , CO, and O_3

Evaluation of CH_4 against ATom

- Using updated halogen chemistry reduces bias in CH_4 compared to Atom measurements
- The same trend exist in different seasons

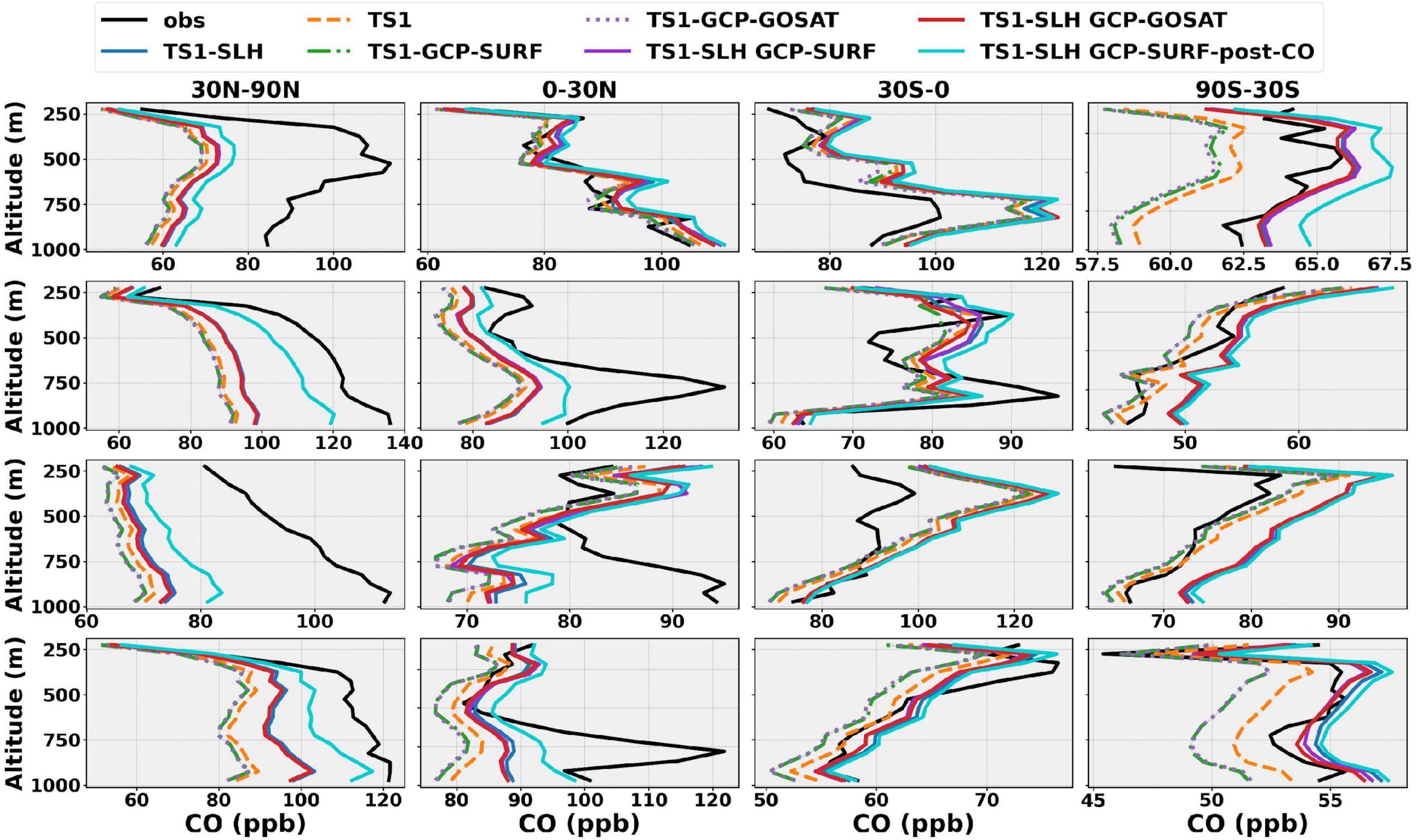
July-Aug	ATom1
Jan-Feb	ATom2
Sep-Oct	ATom3
Apr-May	ATom4



Evaluation of CO against ATom

- Using updated halogen chemistry, and CH_4 emissions reduces bias in CO compared to ATom measurements
- There is smaller variation in CO using updated halogen chemistry and emissions compared to CH_4 variations

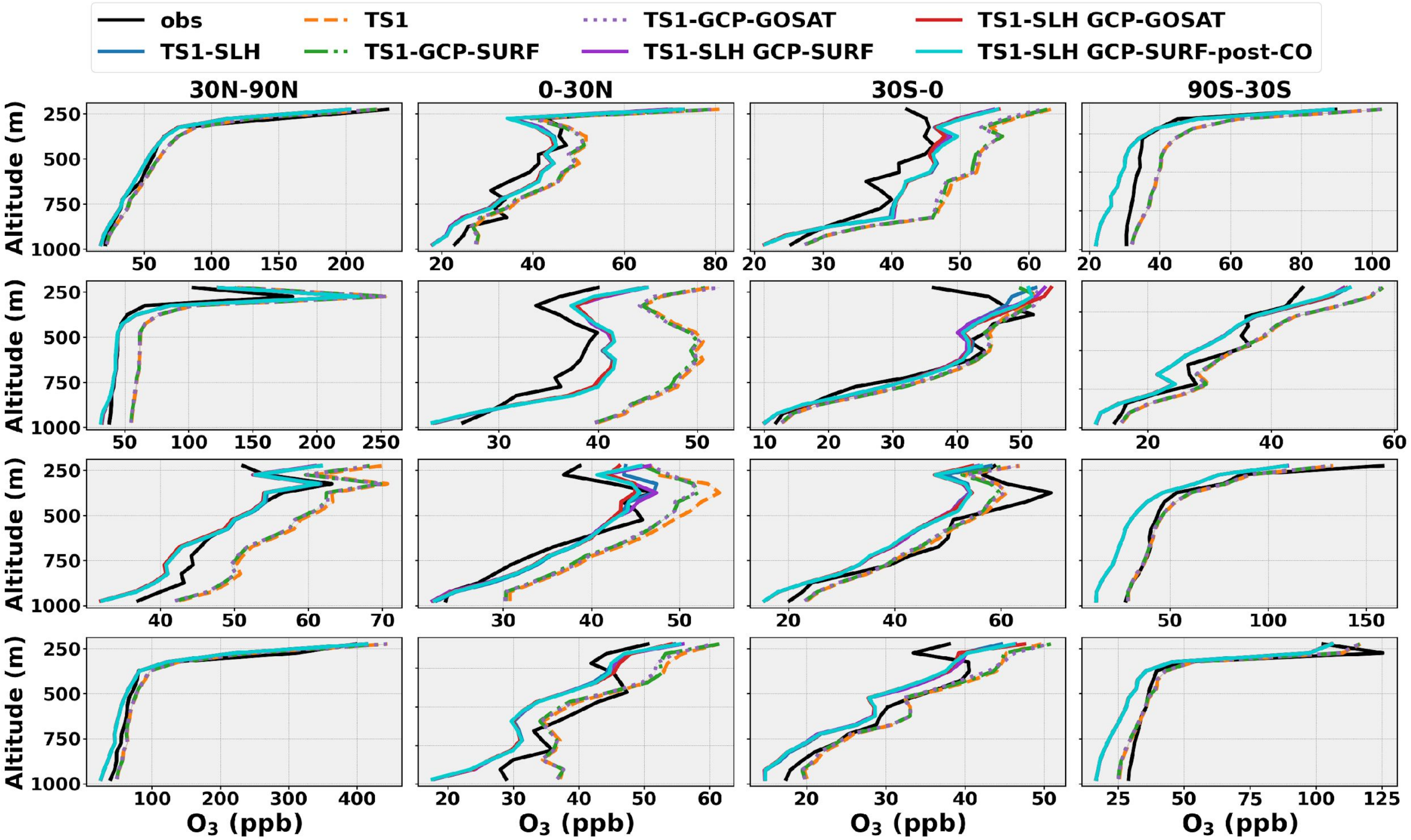
July-Aug	ATom1
Jan-Feb	ATom2
Sep-Oct	ATom3
Apr-May	ATom4



Evaluation of O_3 against ATom

- There is a significant improvement in O_3 using updated halogen chemistry
- The impact of CH_4 and CO emissions is small compared to updated halogen chemistry

July-Aug	ATom1
Jan-Feb	ATom2
Sep-Oct	ATom3
Apr-May	ATom4

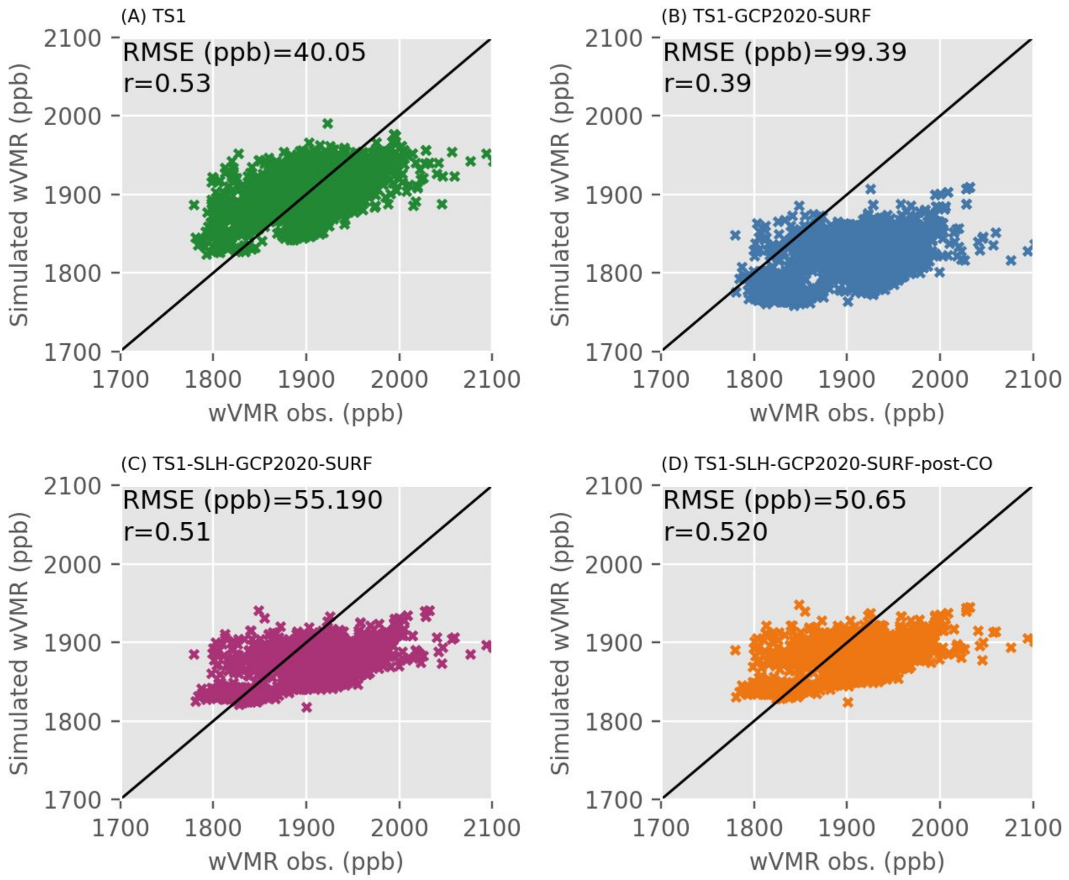


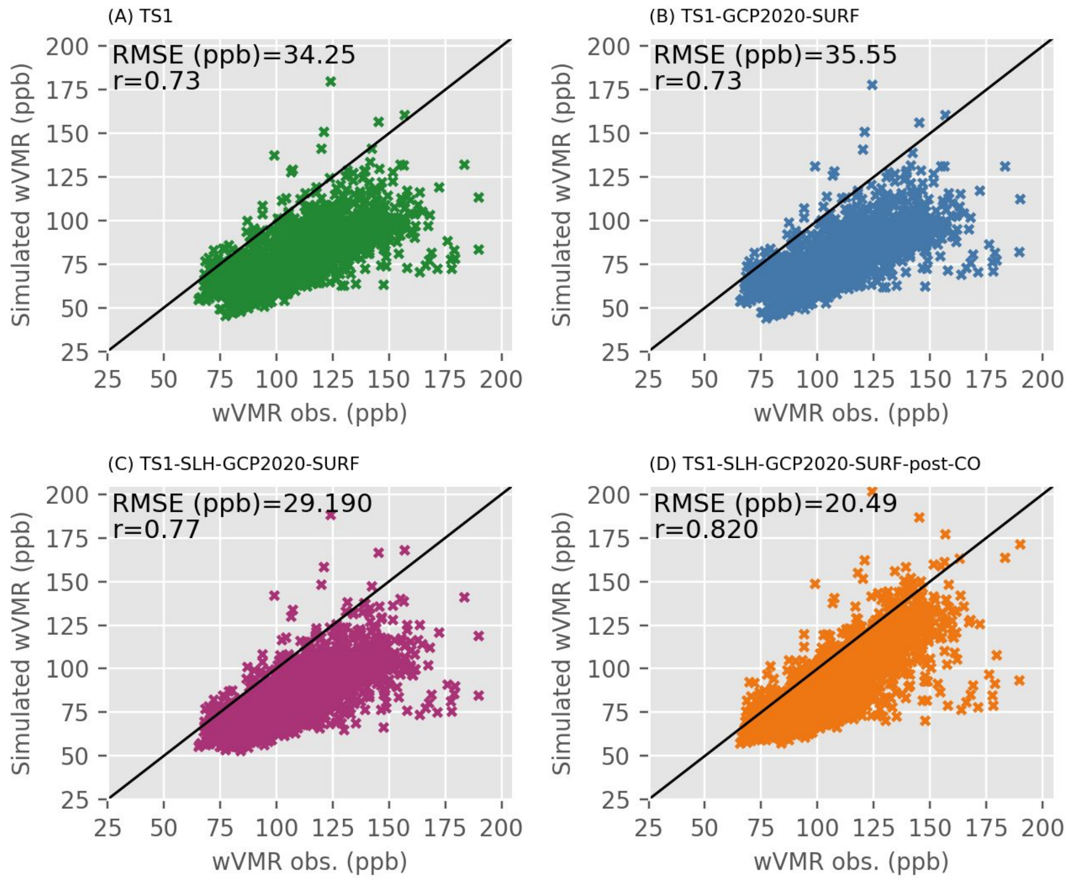
Evaluation of simulations against ATom

Simulation	Mean Bias CH_4 (ppb)	Mean Bias CO (ppb)	Mean Bias O_3 (ppb)	Mean Correlation CH_4	Mean Correlation CO	Mean Correlation O_3
TS1	-1.01	-21.06	9.15	0.94	0.49	0.96
TS1-GCP-SURF	-83	-22.7	8.56	0.96	0.49	0.96
TS1-GCP-GOSAT	-93	-22.25	8.56	0.94	0.51	0.96
TS1-VSL	-0.19	-16.5	-1.92	0.95	0.54	0.95
TS1-VSL GCP--SURF	-32.85	-17.2	-2.1	0.96	0.53	0.95
TS1-VSL GCP-GOSAT	-42.7	-17.7	-2.2	0.952	0.54	0.96
TS1-SLH GCP-SURF-post-CO	-26.34	-11.09	-2.0	0.96	0.63	0.95

Ground-based FTS (NDACC): Northern hemisphere sites

- **Updated halogen representation improves CH_4 and CO**
- **Posterior CO emissions improves CH_4 while emission driven methane simulations improves CO**





Summary

- Using updated halogen chemistry in CESM 2.2 improves CH_4 and CO in the background atmosphere sampled by NASA ATom
- Use of posterior CO emissions improves CH_4
- Halogen representation enables reasonable emission-driven methane simulations
- Sensitivity to the choice of chemistry is larger in emission-driven methane simulations
- Emission-driven methane simulations improves CO

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