Investigating the Role of Short-Lived Halogen Chemistry and Chemical Feedback on CH_4-CO-OH Distributions Using Emission-Driven CESM/CAM-Chem Simulations

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Background

The hydroxyl (OH) radical is the primary oxidant in the Earth's atmosphere, regulating gaseous air pollutants like carbon monoxide and greenhouse gases such as methane.

How does including halogens improve constraints on OH?

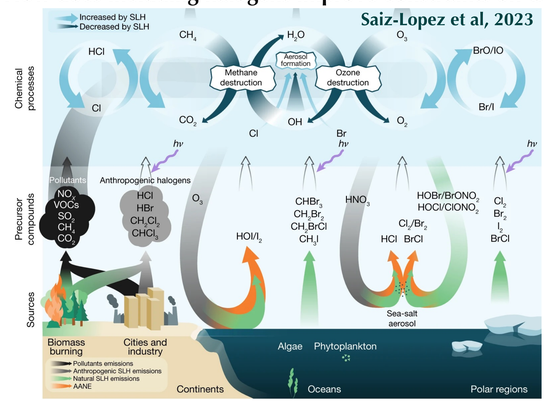


Fig.1 Impact of Short-Lived Halogen compounds on atmospheric oxidation

Research questions and objectives

1. **How does the interplay of OH, CH_4, and CO change in the presence of SLH**
2. **How do Short-Lived Halogens (SLH) affect the abundance and reactivity of tropospheric hydroxyl radicals (OH)?**
3. **What is the impact of halogen-driven changes in OH levels on the lifetime and budget of atmospheric methane (CH_4)?**

We implement an emission-driven configuration in the Community Atmosphere Model with chemistry (CAM-chem) in the **Community Earth System Model Version 2.2 (CESM2.2)** for **2014-2018** period and demonstrate the effect of incorporating detailed **Short-Lived Halogen (SLH)** chemistry representation on both **emission- and concentration-driven** CH_4 simulations

Model Configuration

Simulations	Chemistry	Methane	Anthro CO emis.	Fire CO emis.
TS-CMIP6	TS1.2	CMIP6	CAMS-GLOB-ANT_v5.3	FINN2.5
SLH-CMIP6	SLH			
TS-GCP	TS1.2	GCP-GOSAT		
SLH-GCP	SLH			
TS-CT	TS1.2	CarbonTracker-CH_4	MOPITT inversion	
SLH-CT	SLH		CAMS-GLOB-ANT_v5.3	
SLH-CT-MOPITT			MOPITT inversion	
SLH-CT-MOPITT-depo⬆				
SLH-CT-MOPITT-depo⬆-⚡⬆				

Impact of SLH on OH main chemical sinks (2016-2018)

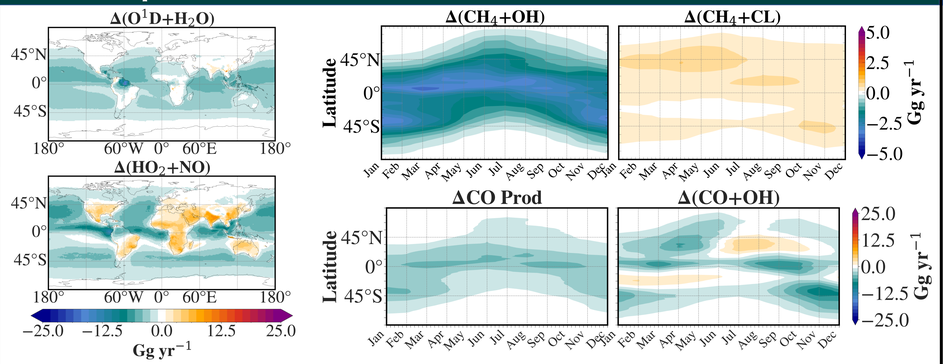
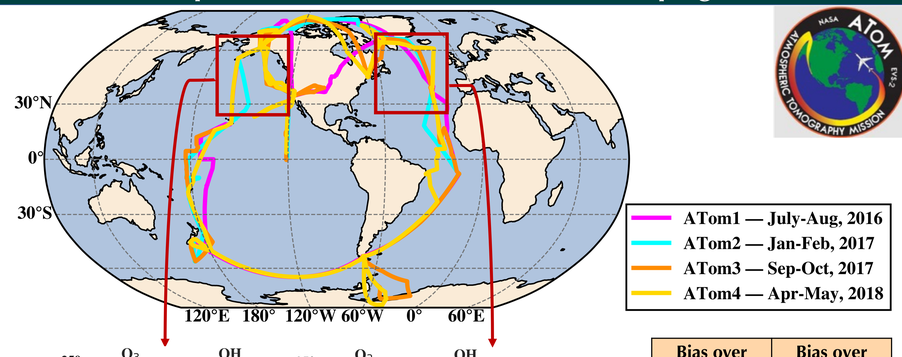
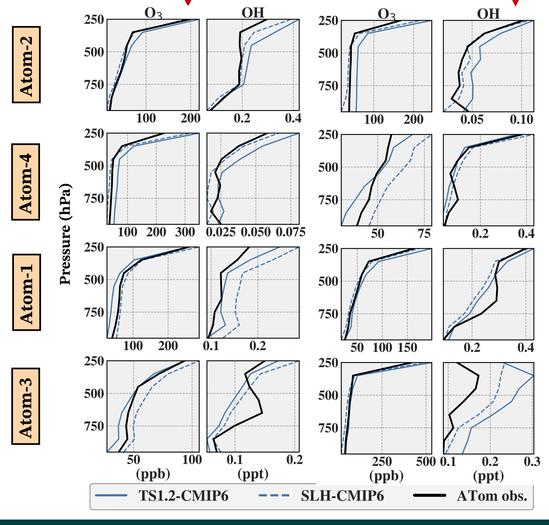


Fig.2 Difference in CH_4 and CO loss and production between simulations with and without Short-Lived Halogen (SLH) representation

- There is a strong **reduction** in **CH_4+OH** loss across the **tropics** and **mid-latitudes.**
- The reduction in OH decreases CO chemical loss and the chemical production during **winter.**
- **CO chemical loss** increases over continental regions in **summer**, probably due to higher OH through the **increase** of **HO_2+NO** secondary OH sources.

Comparison with NASA ATom field campaigns



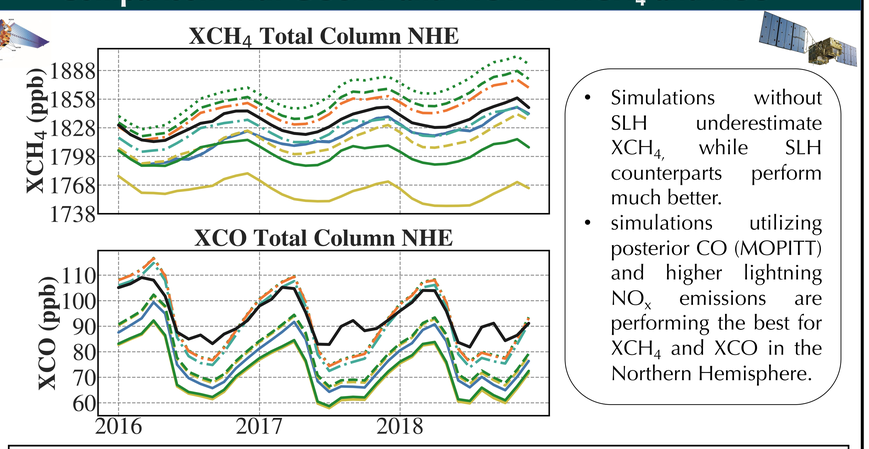


Simulation	Bias over Pacific Ocean >30° O_3 (ppb)	Bias over Pacific Ocean >30° OH (ppq)	Bias over Atlantic Ocean >30° O_3 (ppb)	Bias over Atlantic Ocean >30° OH (ppq)
TS-CMIP6	14.6	28.2	16.6	27.6
SLH-CMIP6	-1.5	3.6	2.3	23.5

- The average bias for the four campaigns in the Northern Hemisphere is considerably reduced.
- Improvement in OH through halogen representation seems to come at the cost of slightly reducing O_3 too much.

Fig.3 Comparison of OH and O_3 vertical profiles with ATom measurements

Comparison with GOSAT and MOPITT XCH_4 and XCO



- Simulations without SLH underestimate XCH_4, while SLH counterparts perform much better.
- simulations utilizing posterior CO (MOPITT) and higher lightning NO_x emissions are performing the best for XCH_4 and XCO in the Northern Hemisphere.

Fig.4 Comparison of CH_4 and CO total columns with GOSAT and MOPITT satellite retrievals

Methane Budget and Lifetime for 2017

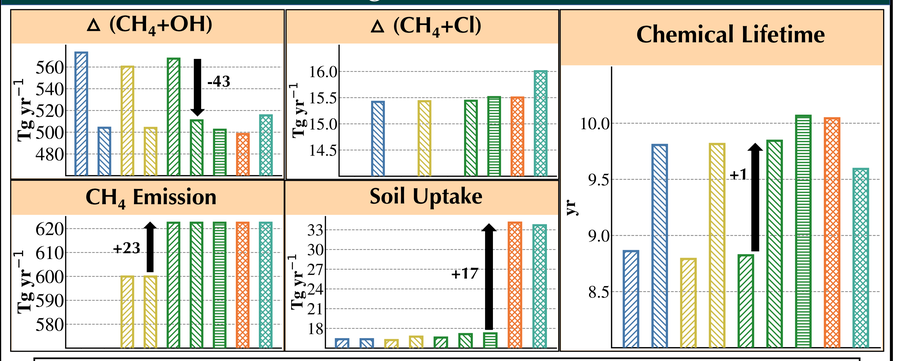


Fig.5 CH_4 annual budget and lifetime for 2017

- The significant reduction in CH_4 oxidation by OH in simulations with the SLH representation is partially compensated by an increase in CH_4+Cl oxidation in the troposphere.
- In emission-driven simulations, the net reduction in CH_4 chemical loss is about 43 $TgCH_4\ yr^{-1}$, with the CH_4 lifetime increasing by one year.
- Chemical loss mechanisms in atmospheric chemistry models are not particularly sensitive to the magnitude of methane emissions.
- Higher lightning emissions result in increased in tropospheric CH_4+OH loss.

Future Work

- Extending simulations to cover longer periods (over a decade).
- Investigating the chemical feedback of CH_4 and CO.
- Joint assimilation of CO and CH_4.
- Evaluating simulations using ground-based measurements and TROPOMI satellite data.

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References

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