Supplemental Information for:

Intended and Unintended Consequences of Atmospheric Methane Oxidation Enhancement

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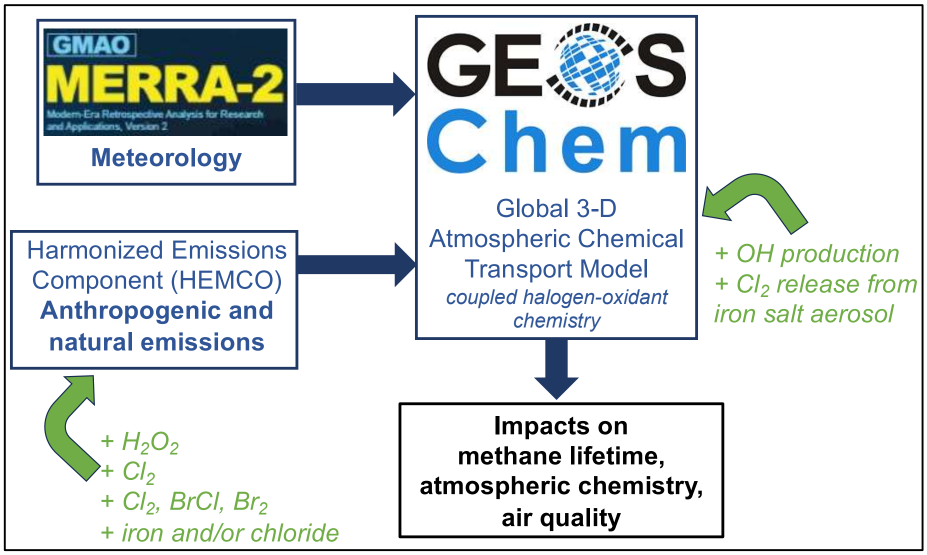

Figure S1. Methodology Schematic.

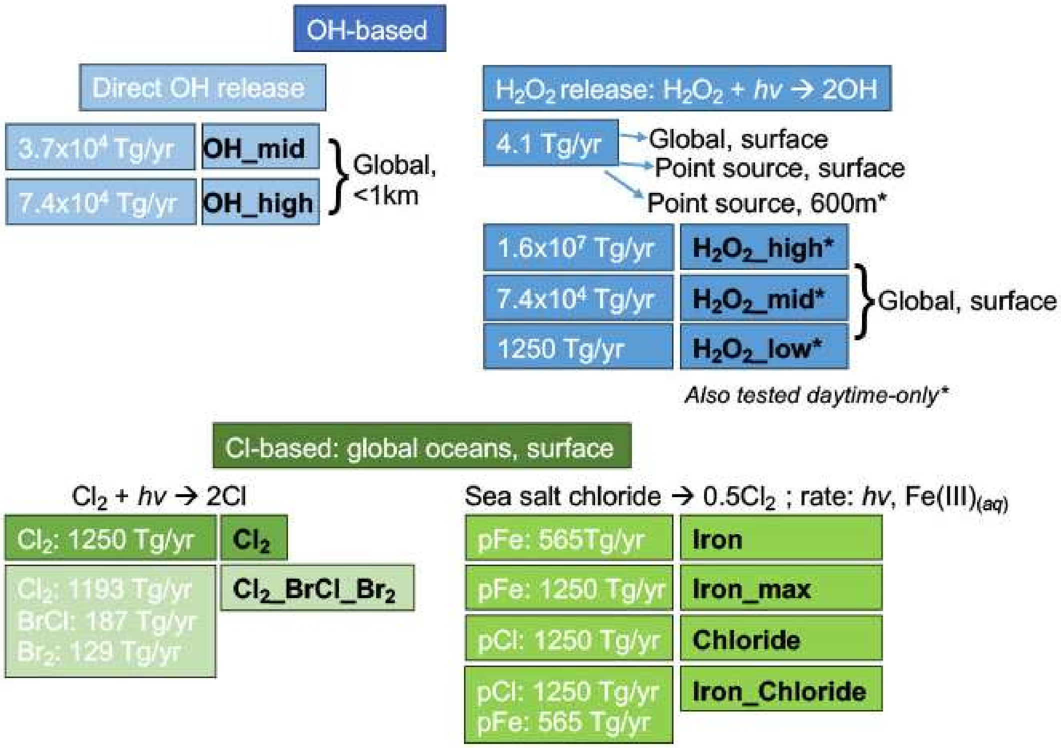

Figure S2. All model experiments conducted in the current study, grouped by methane oxidation mechanism and emitted species.

Table S1 Scenarios with Negligible Results

	Emitted Species	Total Emissions (Tg/yr)	Emissions Location	Emissions Rate at Location of Emissions (kg/m^2/s)
H_2O_2_production	H_2O_2	4.1	Globally at surface	2.549E-13
H_2O_2_point	H_2O_2	4.1	Major natural gas point sources	1.126E-10
H_2O_2_point_600m	H_2O_2	4.1	Major natural gas point sources, at 600 m altitude	1.126E-10; also tested with 2x emissions during daytime only (6am–6pm)

Table S2 Direct OH Release Scenarios

	Emitted Species	Production Rate (Tg/yr)	Emissions Location	Reaction, Rate Coefficient
OH_mid	OH	3.7E4	Globally up to 1 km	$[O_2]$*1.78E-12
OH_high	OH	7.4E4	Globally up to 1 km	$[O_2]$*3.56E-12

Table S3 Comparison of Q. Li et al. (2023) and the Current Study Relevant to the Cl_2 and Cl_2_BrCl_Br_2 Scenarios

	Q. Li et al. (2023)	This Study
Model	CAM4-Chem-CESM-1.1	GEOS-Chem 13.2.1
Model run time	2020–2050	2019
Anthropogenic emissions	Representative concentration pathway 8.5 time-varying	CEDS CMIP6, year 2019
Halogen chemistry	Li et al. (2022); includes Saiz-Lopez et al. (2014) and Ordóñez et al. (2012) plus chlorine and bromine release from sea salt+N_2O_5 (also in GEOS-Chem), sea salt+HNO_3 (not in GEOS-Chem), and N_2O_5+HCl based on Hossaini et al. (2016)	Wang et al. (2021); includes Wang et al. (2019), Sherwen, Evans, et al. (2016), Sherwen, Schmidt, et al. (2016), and Zhu et al. (2019), plus sulfate formation from HOBr and HOCl, N_2O_5+Cl^- from Wang et al. (2020), improved reactive uptake coefficients on ice crystals for halogens
Model resolution	1.9° × 2.5° horizontal	4° × 5° horizontal
Model resolution	26 vertical levels up to 40 km	72 vertical layers up to 80 km; chemistry performed in first 59 layers up to 49.8 km

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Table S4 Comparison of van Herpen et al. (2023) and Current Study for Iron and Chloride Scenarios

	van Herpen et al. (2023)	This Study
Model	CAM4-Chem	GEOS-Chem 13.2.1
Model run time	June 1996–1998	2019
Aerosols participating	Dust 1–2.5 μm	Dust 0.1–1 μm; Accumulation mode sea salt chloride (≤0.5 μm)
Dust iron content	3.5%	3.5%
Photoactive iron	Dust: 1.8%	Dust: 2.68% Anthropogenic: 26.8%
Reaction rate for Cl_2 release	Fe(II)–Fe(III) cycling kinetics from Zhu et al. (1993) (calculations based on field samples)	Based on Wittmer, Bleicher, and Zetzsch (2015) chamber study (Chen et al., 2024)
Halogen chemistry	Wang et al. (2021)	Wang et al. (2021)
Horizontal model resolution	0.9° × 1.25°	4° × 5°
Vertical model resolution	56 vertical levels up to 40 km	72 vertical levels up to 80 km; chemistry performed in first 59 layers up to 49.8 km

Table S5 Changes in Tropospheric Burdens for the OH Experiments

	Br_y	Cl_y	I_y	O_3	OH	Cl	NO_x	CO
Standard	20 Gg	241 Gg	12 Gg	338 Tg	215 Mg	318 kg	359 Gg	349 Tg
OH_mid	41.2	11.6	-17.8	-10.4	50.8	45.2	-21.1	-26.4
OH_high	54.6	19.5	-28.3	-14.3	94	72.3	-24.7	-31.1

TABLE S6 Percent Change in Tropospheric Burdens of Other Climate Forcers and Ozone-Depleting Substances, with Cl Atom Included for Reference, for Experiments Described in Table 1

	HCFC-123	Total inorganic aerosol	Dibromo-methane (CH_2Br_2)	Bromoform ($CHBr_3$)	Methyl ioidide (CH_3I)	Cl	ozone
H2O2_high	-74.2	11.0	-61.0	-39.3	-6.6	396.4	-38.5
H2O2_mid	-34.7	13.7	-30.7	-20.9	-3.0	75.2	-6.3
H2O2_low	-3.8	7.3	-3.8	-2.6	-0.4	6.5	-0.8
Cl2	30.7	6.8	36.9	10.8	-1.1	2185.2	-24.4
Cl2_BrCl_Br2	64.0	20.9	76.2	12.1	-7.9	1839.8	-67.7
Iron	2.7	10.0	2.8	1.0	-0.2	179.3	-3.5
Chloride	5.3	-0.5	5.6	2.3	-0.2	180.9	-5.5
Iron_Chloride	11.0	10.9	11.8	4.4	-0.5	680.8	-10.7

TABLE S7 Percent Change in Stratospheric Burdens (tropopause up to 50 km) of Other Climate Forcers and Ozone-Depleting Substances, with Cl Atom Included for Reference, for Experiments Described in Table 1

	HCFC-123	Total inorganic aerosol	Dibromo-methane (CH_2Br_2)	Bromoform ($CHBr_3$)	Methyl ioidide (CH_3I)	Cl	ozone
H2O2_high	-0.7	-2.0	-61.0	-60.6	-11.0	0.3	-0.3
H2O2_mid	-0.3	-1.3	-27.0	-23.4	-3.9	0.0	-0.1
H2O2_low	0.0	-0.2	-3.1	-2.5	-0.5	0.0	0.0
Cl2	0.3	3.4	36.9	16.0	-2.4	-3.1	-0.5
Cl2_BrCl_Br2	0.5	9.2	76.2	11.7	-15.6	-1.5	-7.8
Iron	0.0	0.4	2.5	1.1	-0.4	0.0	0.0
Chloride	0.0	0.2	5.0	2.4	-0.5	-2.3	-0.3
Iron_Chloride	0.1	1.0	10.9	4.9	-1.1	-2.2	-0.5

TABLE S8 Changes in Tropospheric Species in Reference to Impacts from Future Hydrogen Economy

	OH	Methane	CO	Ozone	NO	Sulfate
Hydrogen[a]	↓	↑	↑	↑	N/A	N/A; shift from new particle formation to particle growth[b]
H_2O_2-based oxidation enhancement[c]	↑	↓	↓	↓	↓	↑
Chlorine-based oxidation enhancement[c]	↓	↓	↑	↓	↓	↑

NOTE: "Hydrogen" here does not include reductions in fossil fuel emissions replaced by hydrogen technology.

[a] Warwick et al., 2023.

[b] O'Connor et al., 2022.

[c] This work.

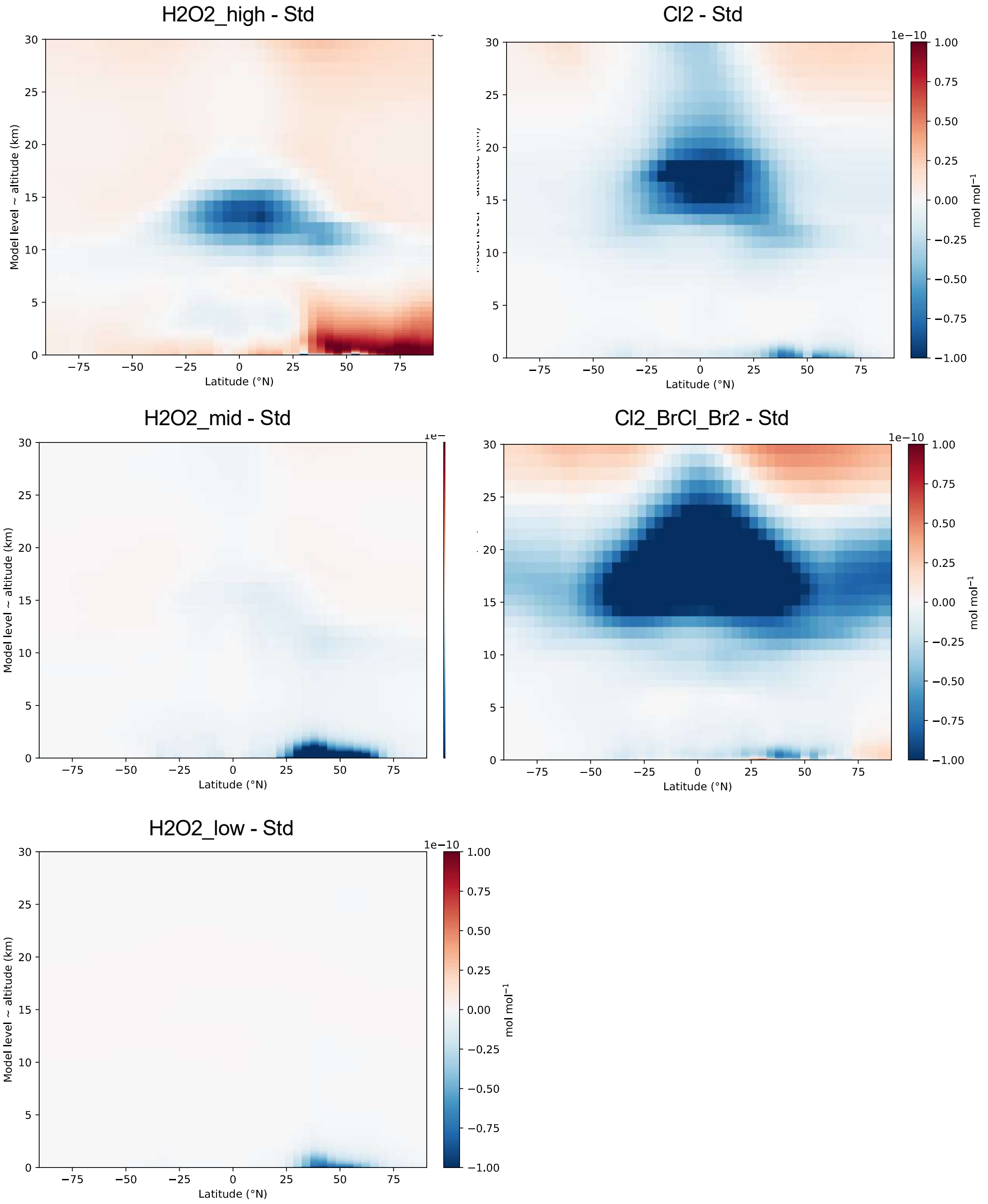

Figure S3 Zonal mean absolute changes in NO_x across the scenarios in mol/mol.

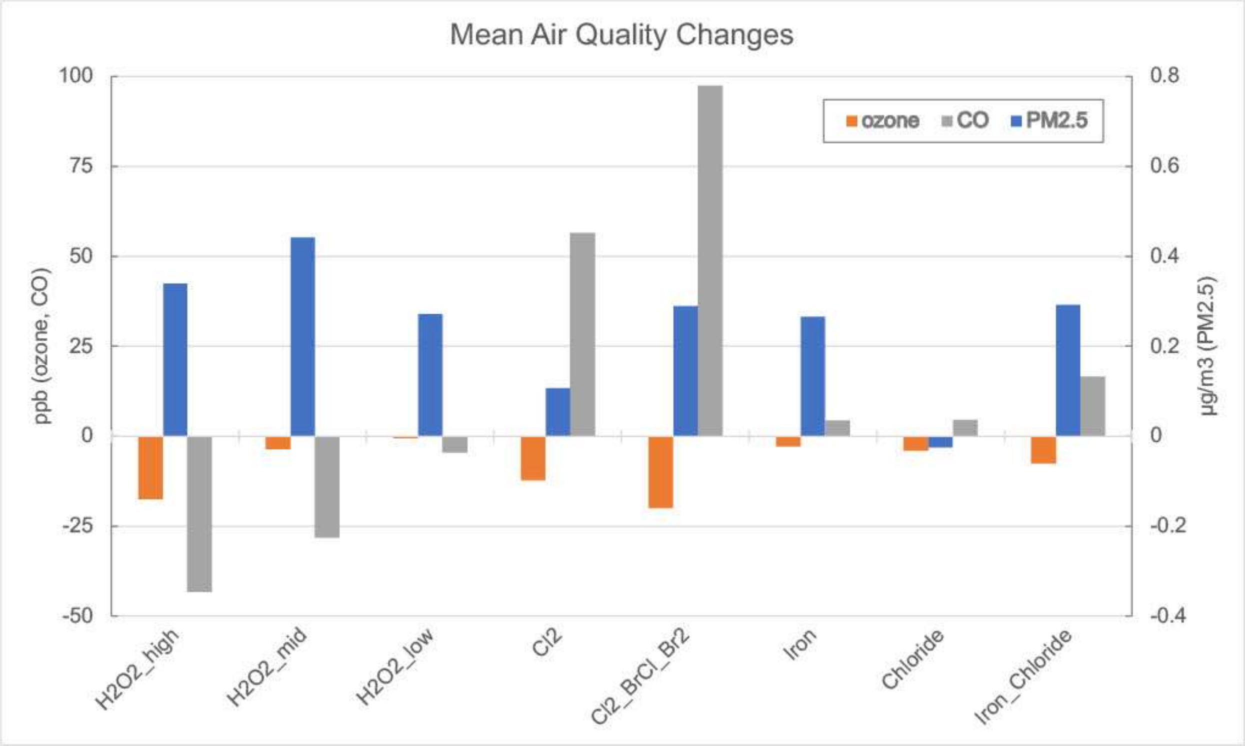

Figure S4 Changes in global annual mean ozone (orange), CO (gray) (left axis), and $PM_{2.5}$ (blue, right axis) concentrations across the experiments.