PREINDUSTRIAL TO PRESENT DAY CHANGES IN TROPOSPHERIC HYDROXYL FROM ACCMIP

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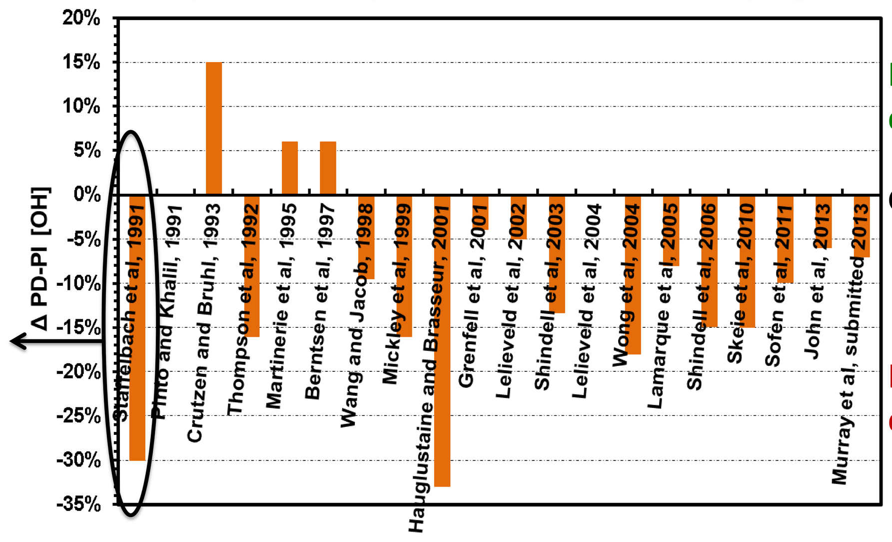
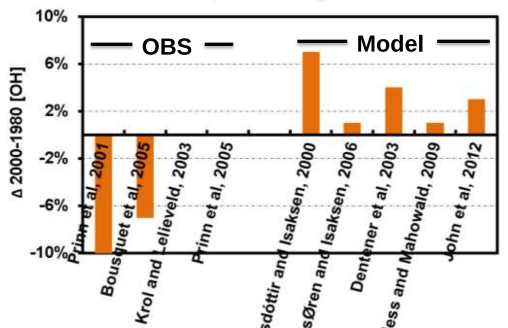
1. Motivation

❖ Accurate measurements and modeling of tropospheric hydroxyl (OH) are key to understanding photochemical oxidation and better quantify the lifetimes of climate relevant gases, such as methane (CH_4), hydrochlorofluorocarbons (HCFCs) and hydrofluorocarbons (HFCs)

❖ Based on previous literature, the extent to which global mean tropospheric OH has changed from preindustrial (1850) to present day (2000) due to anthropogenic activity is highly uncertain.



❖ OH change from 1980 to 2000 inferred from methyl chloroform (CH_3CCl_3) measurements in previous studies is inconsistent with that simulated by models. However, recent analysis of CH_3CCl_3 data post 1997 indicates that global OH is generally well-buffered against perturbations [Montzka et al., 2011; Holmes et al., 2013].



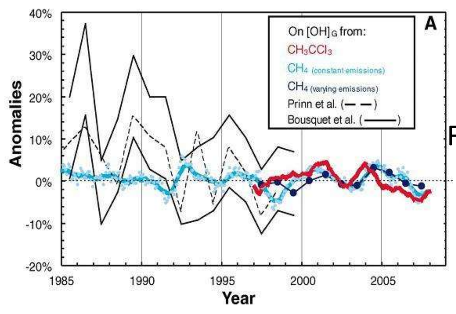


Figure from Montzka et al. [2011]

Objectives

❖ **Evaluate present day global OH in current generation chemistry-climate models (CCMs)**

❖ **Explore changes in OH and methane lifetime for present day (2000) relative to preindustrial (1850) and to 1980**

❖ **Investigate the impact of individual factors (climate and emissions) in driving preindustrial to present day OH changes**

2. The Atmospheric Chemistry & Climate Model Intercomparison Project (ACCMIP)

CESM-CAM-superfast (CE)	LLNL-NCAR, USA
CICERO-OsloCTM2 (CI)*	CICERO, Norway
CMAM (CM)	CCCma, Canada
GEOSCCM (GE)	NASA GSFC, USA
GFDL-AM3 (GF)	NOAA GFDL, USA
GISS-E2-R (GI)	NASA GISS, USA
GISS-E2-R-TOMAS (GT)	NASA GISS, USA
HadGEM2 (HA)	UKMO, UK
LMDzORINCA (LM)	LSCE, France
MIROC-CHEM (MI)	NIES, Japan
MOCAGE (MO)*	MeteoFrance, France
NCAR-CAM3.5 (NC)	NCAR, USA
STOC-HadAM3 (ST)	Univ. Edinburgh, UK
TM5 (TM)*	RNMI, The Netherlands
UM-CAM (UM)	NIWA, New Zealand

* CTMs

❖ CCMs mostly driven by SST/SIC from parent coupled models

❖ WMGHGs mostly from Meinshausen et al. [2011], CH_4 concentrations specified at the surface in most models

❖ Anthropogenic emissions from Lamarque et al. [2010], diverse natural emissions

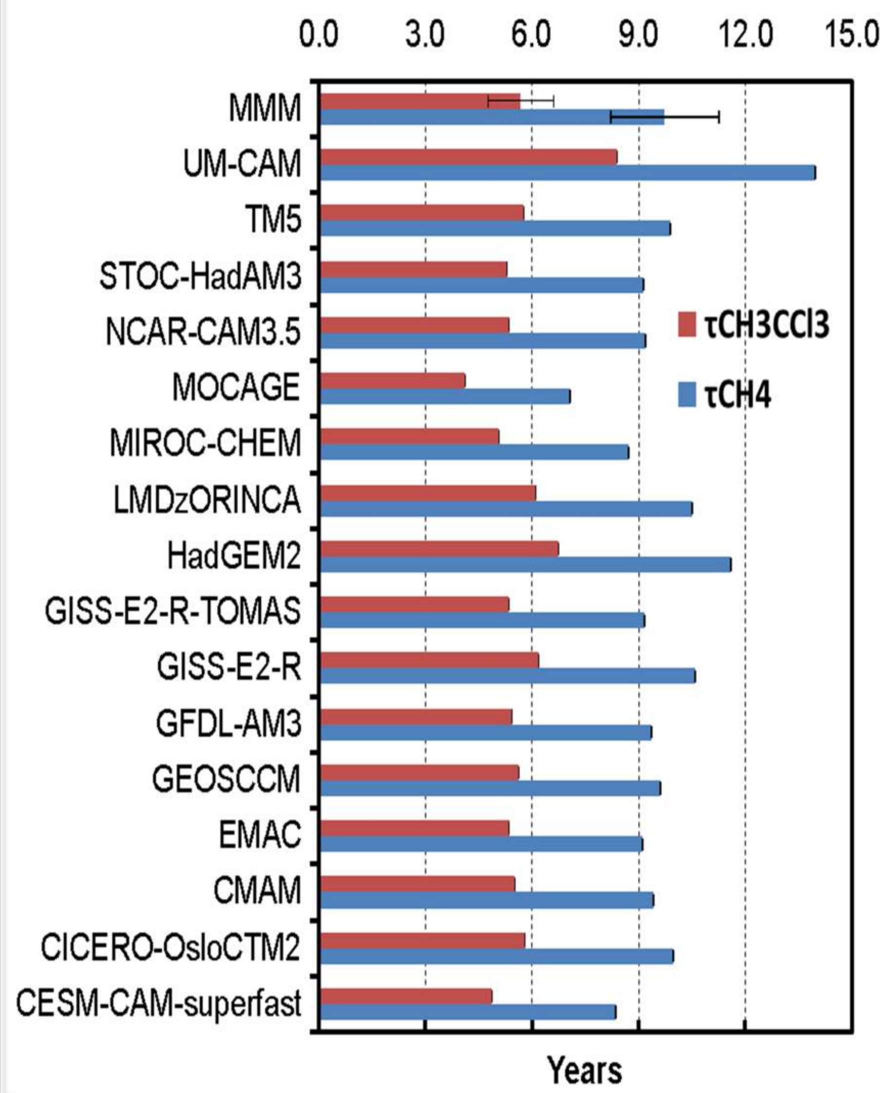
❖ Diverse representation of stratospheric O_3 and its influence on photolysis, and tropospheric chemistry mechanisms

❖ Time slice simulations (run for ~4 to 10 years): 1850, 1980, 2000

❖ Fixed 2000 emission and 1850 climate

❖ Fixed 2000 climate with CH_4 conc. and NO_x, CO, NMVOCs emissions set to 1850 individually

Present day Tropospheric CH_4 Lifetime: Large spread attributed to model-to-model differences in NMVOC emissions and simulated ozone photolysis [Voulgarakis et al., 2013]



Present day multimodel mean (MMM) τ_{CH4} and $\tau_{CH3CCl3}$ are 5-10% lower than observation-based estimates but within the uncertainty range

MMM $\tau_{CH_3CCl_3}$ = 5.7±0.9 years

Prinn et al. [2005] $\tau_{CH_3CCl_3} = 6.0^{+0.5}_{-0.4}$

Prather et al. [2012] $\tau_{CH_3CCl_3}$ = 6.3±0.4

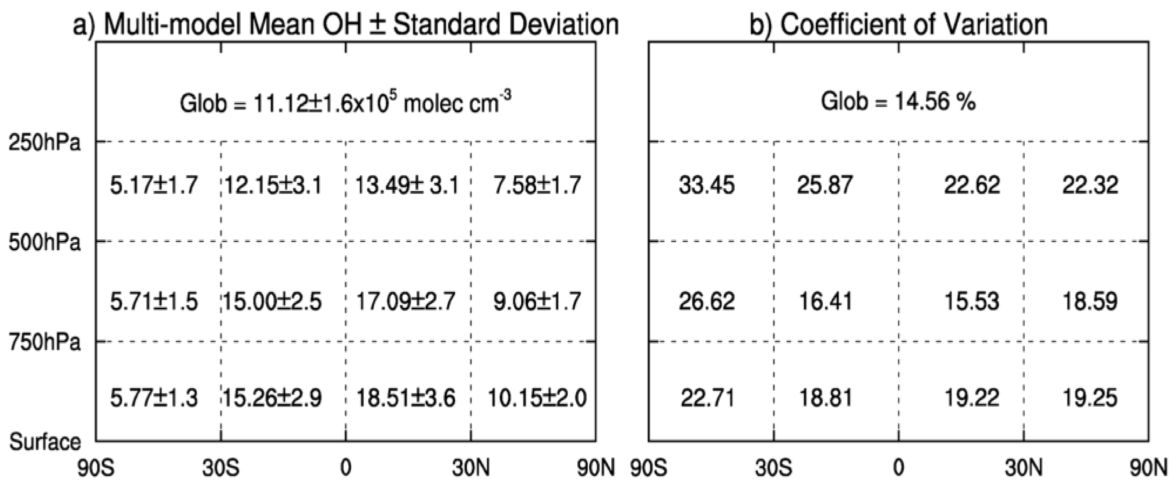
MMM τ_{CH_4} = 9.7±1.5 years

Prinn et al. [2005] $\tau_{CH_4} = 10.2^{+0.9}_{-0.7}$

Prather et al. [2012] τ_{CH_4} = 11.2 ± 1.3

MMM [OH] = 11.1±1.6 x 10^5 molec cm^{-3} is overestimated by 5-10% but is within the range of uncertainties

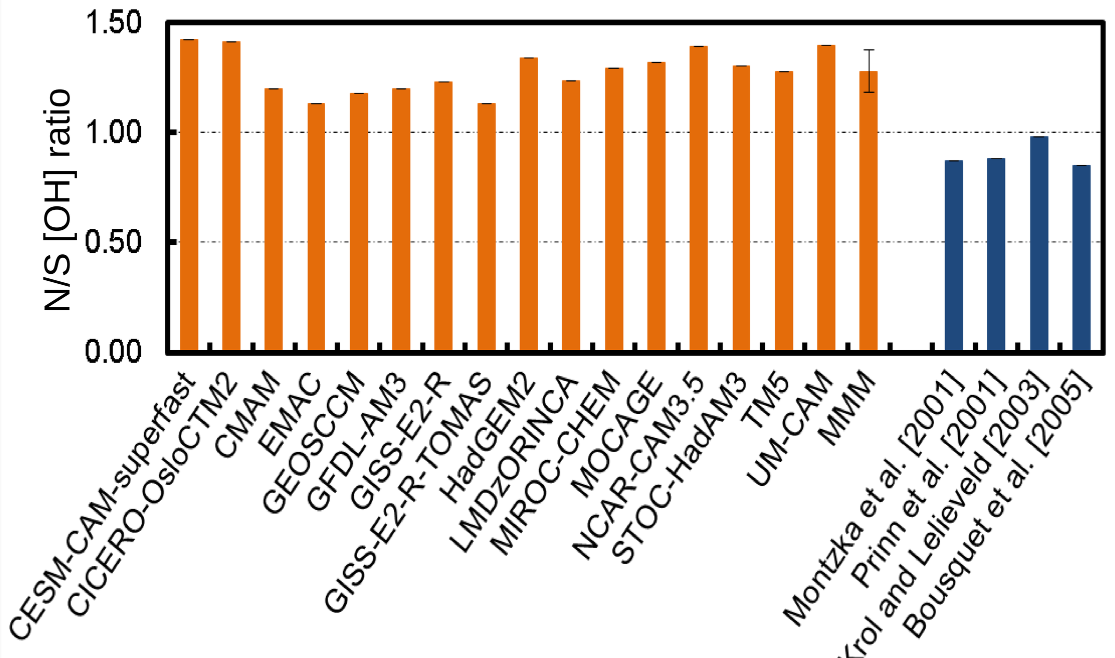
3. Regional Distribution of Present day OH: Large spread but models capture the general spatial patterns



❖ Greater diversity in upper troposphere than in mid to lower troposphere possibly reflecting biases in water vapor and clouds

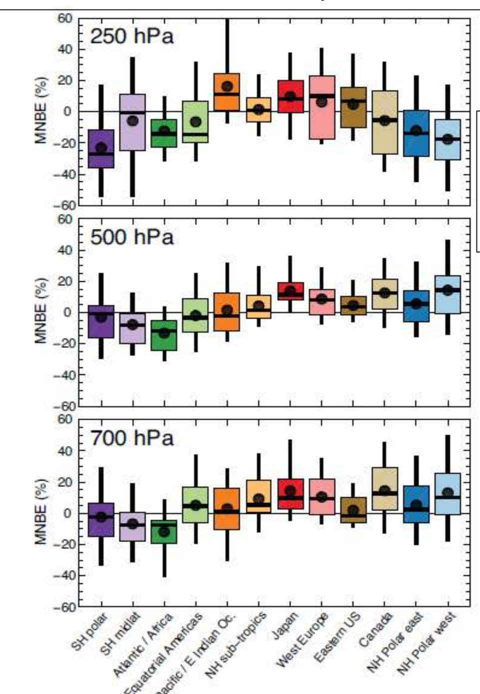
❖ Diversity is also greater in the Southern Hemisphere (SH) than the Northern Hemisphere (NH) reflecting the intermodel diversity in simulated (or prescribed) stratospheric ozone and its influence on tropospheric ozone photolysis

All models have greater OH in the Northern Hemisphere (NH) than the Southern Hemisphere (SH)



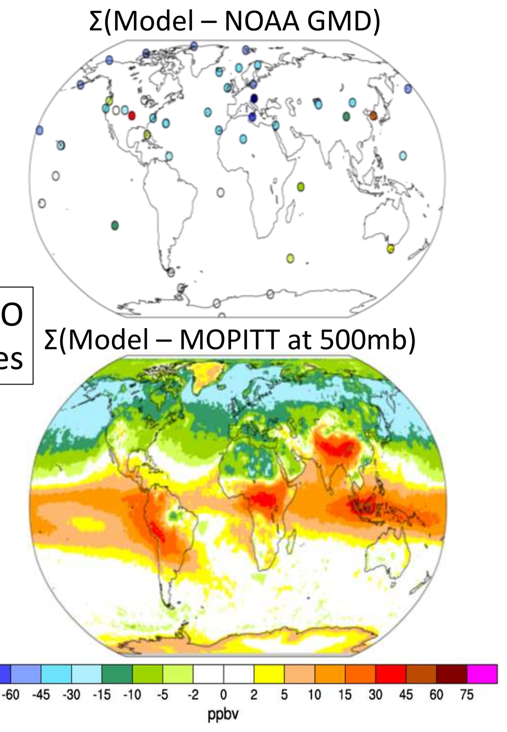
Observational estimates indicate N/S < 1 with 15-30% uncertainties

Present day Model O_3 versus Ozonesonde measurements (1995-2009)



Positive NH tropospheric O_3 (OH source) biases

Present day Model CO versus Measurements



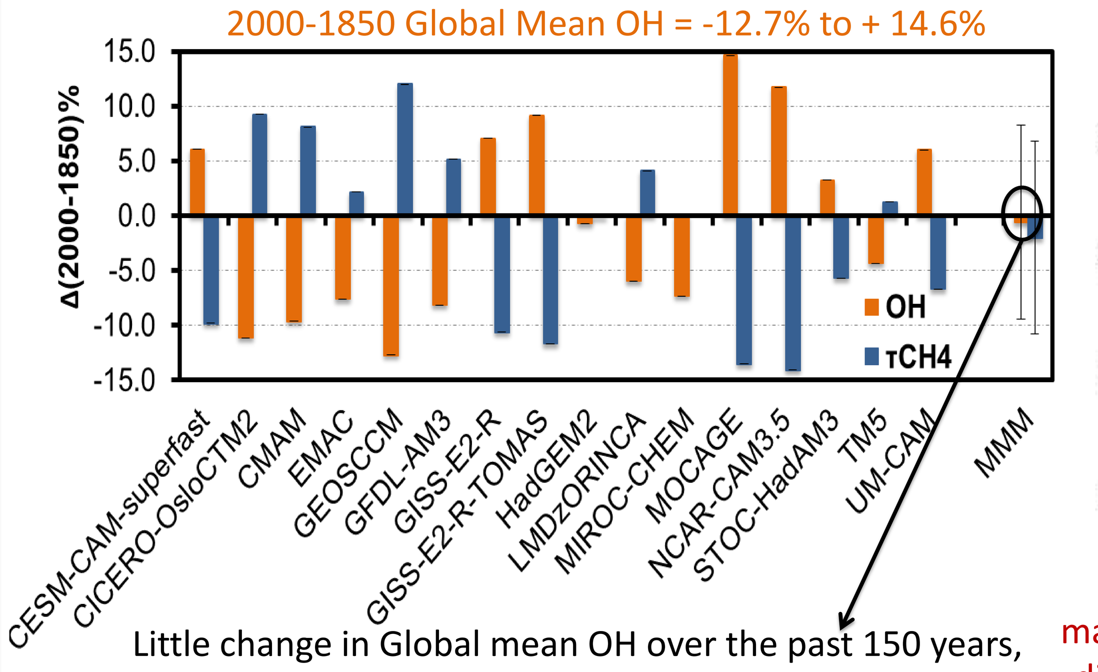
Negative NH CO (OH sink) biases

Consistent with larger OH north-to-south asymmetry (N/S>1)

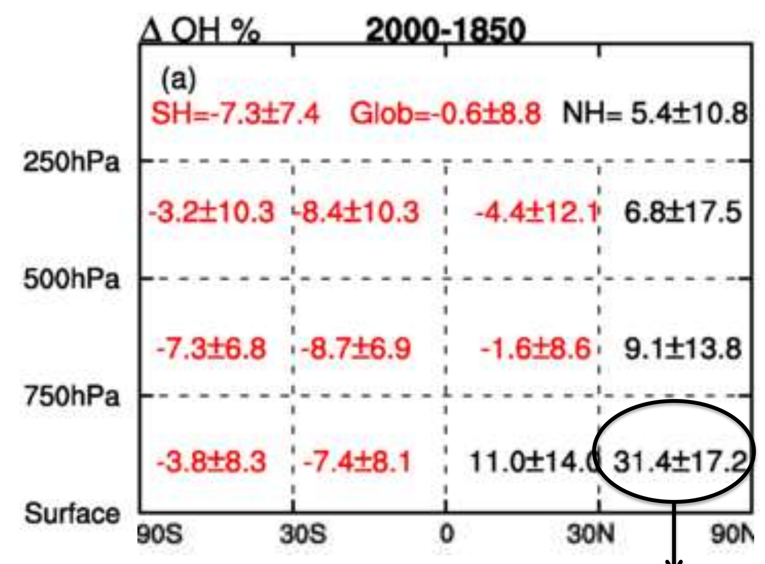
Figure from Young et al. [2013]

4. Present day OH relative to preindustrial

2000-1850 Global Mean OH = -12.7% to + 14.6%



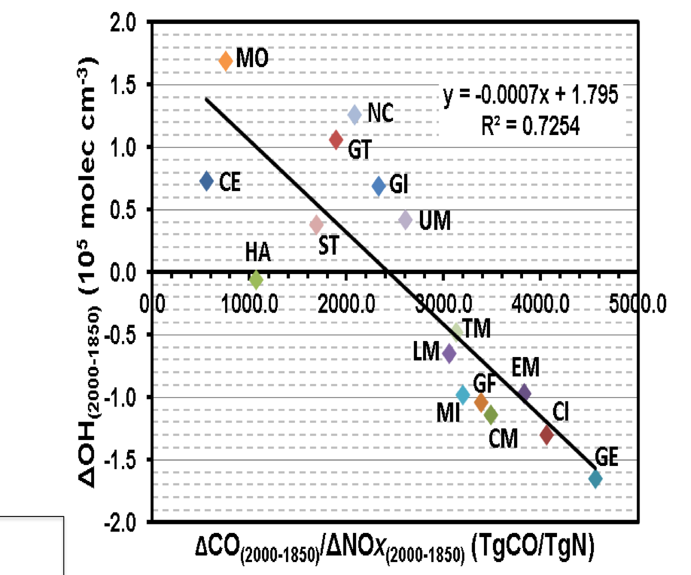
Little change in Global mean OH over the past 150 years, indicates that it is well-buffered against perturbations [Lelieveld et al., 2004; Montzka et al., 2011]



Large uncertainties exist in the magnitude and sign of OH change in different atmospheric subdomains

↑NO_x, O_3, H_2O > ↑CO, NMVOCs, CH_4

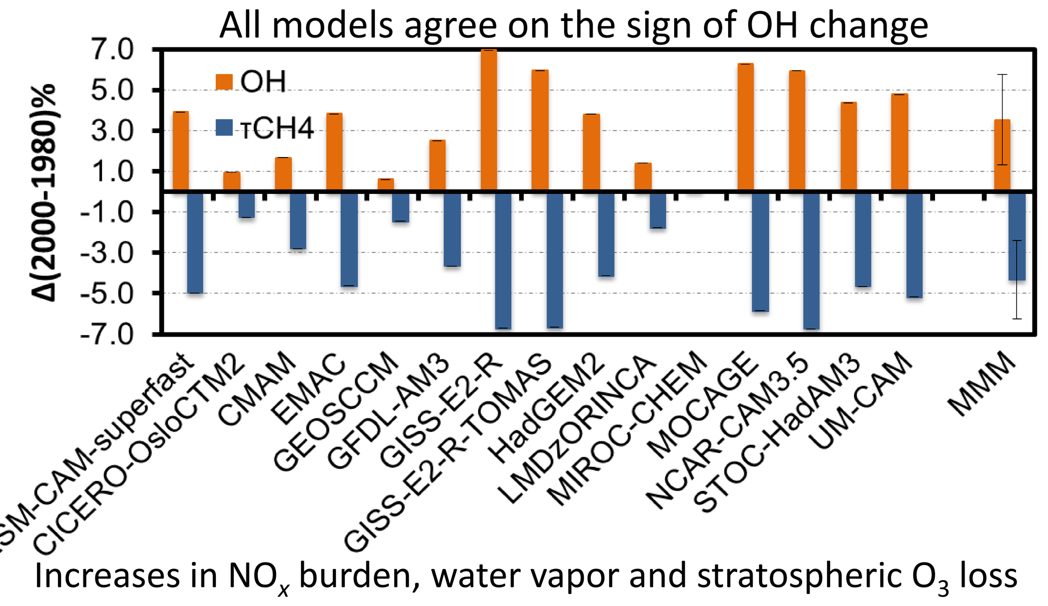
Different $\Delta CO/\Delta NO_x$ (~changes in OH sinks versus sources) drive diverse preindustrial to present day OH changes across models



Uncertainties in natural emissions and chemical mechanisms contribute to the spread in the interplay between OH sources and sinks

5. Present day OH relative to 1980

All models agree on the sign of OH change



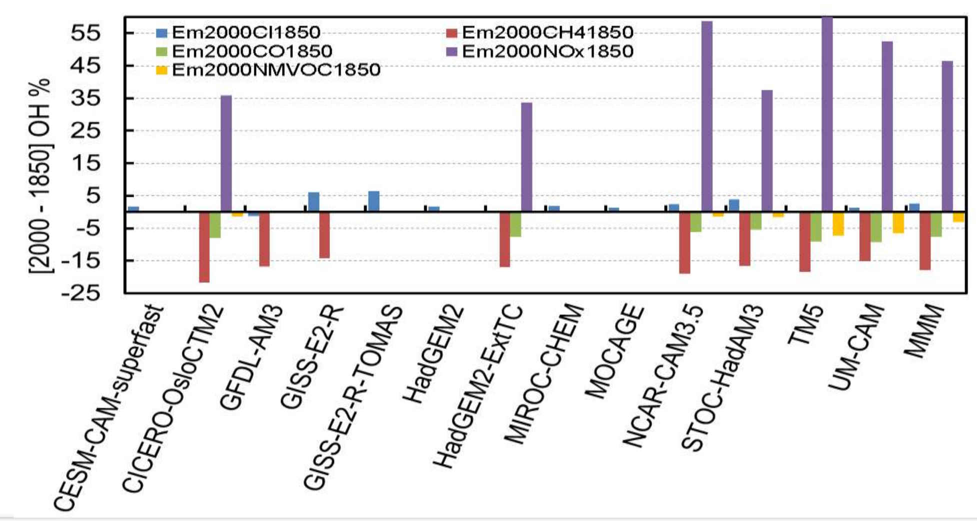
Increases in NO_x burden, water vapor and stratospheric O_3 loss outweigh those in CH_4 and CO burdens resulting in small OH increase consistent with recent estimates [Montzka et al,. 2011; Monteil et al., 2011]

6. Impact of Climate Change on Tropospheric CH_4 Lifetime: τ_{CH4} decreases by about 4 months

Warmer temperatures and higher humidity levels enhance OH-induced loss of CH_4 thereby decreasing its lifetime and contributing a negative climate feedback, consistent with previous studies [e.g. Stevenson et al., 2000]

Models	$\Delta\tau_{CH4}$ (years)	ΔT (K)	$\Delta\tau_{CH4}/\Delta T$ (years K^{-1})
CESM-CAM-superfast	-0.27	1.4	-0.20
GFDL-AM3	0.12	0.6	0.21
GISS-E2-R	-0.76	1.1	-0.69
GISS-E2-R-TOMAS	-0.70	1.1	-0.64
HadGEM2	-0.20	0.5	-0.40
MIROC-CHEM	-0.25	0.8	-0.30
MOCAGE	-0.20	0.9	-0.23
NCAR-CAM3.5	-0.37	1.1	-0.34
STOC-HadAM3	-0.46	0.6	-0.71
UM-CAM	-0.34	0.6	-0.55
MMM±STD	-0.30±0.24	0.9±0.3	-0.39±0.28

7. Influence of changes in CH_4 and anthropogenic emissions on PI to PD OH changes: NO_x > CH_4 > CO > NMVOCs



8. Future Considerations

❖ Accurate observational constraints on OH either through direct measurements or using proxies are needed to improve our understanding of OH as represented in models

❖ Questions remain about the role of aerosols (via chemistry), clouds, NMVOCs (OH recycling in low NOx, biogenic emission changes), and lightning in driving OH changes

9. References

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Gordon Research Conference on Atmospheric Chemistry, July 28 – August 2, 2013