Changes in Global Tropospheric OH Expected as a Result of Climate Change Over the Last Several Decades


1University of Maryland, College Park, Md; 2NASA Goddard Space Flight Center, Greenbelt, Md; 3Johns Hopkins University, Baltimore, Md; 4Universities Space Research Association, Columbia, Md

1. Scientific motivation for studying tropospheric OH

Sensitivity of OHTROP to climate change

- Best knowledge of global tropospheric OH (OHTROP) comes from analysis of CH4, CO2 observations.
- The CH4 lifetime inferred from OHTROP based on CH4CL is 8.9 years; yet many sources use a CH4 lifetime of ~12 years, which was found by modeling studies described in IPCC (2001).
- The 12 year CH4 lifetime is central to subsequent IPCC reports and is called the “perturbation lifetime.”
- Our work is motivated by understanding what factors other than rising CH4 will affect OHTROP.

Methane Oxidation and OH widening

- Under low NOx, the oxidation of CH4 destroys NOx.
- Under high NOx, the oxidation of CH4 produces NOx.
- Loss of methlyperoxy radical CH3O2 with NO or HO2 is critical.
- Future levels of CH4 and NOx are highly uncertain. CH4 between now and 2100 varies dramatically among IPCC scenarios.
- NOx between now and 2100 will depend on whether the developing world implements selective catalytic reduction on coal power plants and catalytic converters on cars.
- Future OHTROP will also depend on overhead column O3, local humidity, and biogenic emission of VOCs, etc.
- Given this complexity, we expect CCMs to project a wide range of values for OHTROP during the rest of this century.

Importance of OHTROP

- OH serves as the main sink for many species: CO, CH3, CH3COCl, all HFCs and HFCS, biogenic VOCs (isoprene and terpenes), anthropogenic VOCs (formaldehyde, benzene, toluene, etc.)
- NOx is integral to O3 formation and destruction pathways; alone, NOx depletes O3, but with NOx and VOCs, HO2 creates O3.

2. Methods for estimating changes in OHTROP

Model OH

- Initial modern-day conditions for OH are taken from a time-slice run of GECOS CCM using 2006 emissions.
- Monthly mean mixing ratios of OH and related species are provided on a 2.5° by 2.5° lat/lon grid.
- Changes in OH due to CO2, overhead O3, and tropical widening are found from analysis of initial OH field.

Overhead O3

- Total column O3 trends were obtained from the NASA merged O3 data set, consisting of measurements from SBUV, TOMS, and Aura OMI instruments.
- We then use our photochemistry code to estimate the impact on OH due to decreases in OH.
- O3 columns by amount suggested by the NASA product.

Tropical Widening

- Widespread evidence for expansion in the OH tropics due to changes in OH.
- The determined scaling factor would be used to calculate a new OHTROP based on the H2O trends.

3. Preliminary results for expected changes in OHTROP

Previous Work on OHTROP Trends

- Montzka et al., 2011 found that OHTROP does not vary interannually (from 1997 to present).
- Prior studies by Prinn et al., 2001 and Bouquie et al., 2005 suggest large interannual variability in OHTROP (1985-2000).
- We suggest the OHTROP behavior in all three studies may be physically possible, based on our preliminary results.

Current Conclusions from OHTROP Analysis

- Effect of CH4 on OHTROP is taken from IPCC (2001), Section 4.2.1.1, which states that “the feedback of CH4 on tropospheric OH found using contemporary chemical transport models is ~0.32% for every 1% increase in CH4 (red line).”
- Primary effect of overhead O3 is rise in OHTROP following the 1995 eruption of Mount Pinatubo (green dashed line) due to enhanced removal of stratospheric O3 by volcanic aerosol.
- Rising H2O from MERRA and AIRS increases OHTROP (orange lines) by an amount comparable to the decrease expected from rising CH4.
- Widening of the tropics contributes an increase in OHTROP (purple line) roughly equal to that due to rising H2O, leading us to view this as a significant factor controlling OHTROP.
- Overall expected change in OHTROP (bottom panel) shows higher level of interannual variability prior to ~1999 and lower variability thereafter.

4. Future plans for refining estimates of ΔOHTROP

Improve Estimate of d(OHTROP) / d(H2O)

- Reaction rates from recent runs of GEOS CCM are archived for reactions such as H2O + O(1D)
- Using these reaction rates we will determine the relationship between CH4 and OHTROP and its dependence on NOx.

Evaluate CH4/OH Feedback

- We will use a box model (details below) to probe sensitivity of OHTROP to changes in CH4.
- We will use a box model (details below) to probe sensitivity of OHTROP to changes in CH4.
- Monthly mean mixing ratios of OH and related species are provided on a 144 longitude, 91 latitude, 72 pressure level grid.
- Changes in OH due to CO2, overhead O3, and tropical widening are found from analysis of initial OH field.

Propagate Uncertainties

- Calculate uncertainties in AIRS and MERRA H2O and NASA O3 product.
- Evaluate standard deviation in average fraction of OH production occurring via H2O + O(1D).
- Estimate uncertainty in the box model evaluation of d(OHTROP) / d(CH4).

Assessing OHTROP in CCMs

- Through our involvement with the IGAC / SPARC Chemistry-Climate Model Initiative, we have requested:
  1. Hourly, instantaneous output from participating CCMs 1 day/season, 1 year/decade
  2. archival of all species, reaction rates, J-values, and physical parameters relevant to OH chemistry
  3. this output for both the REF-CL1 (hindcast) and REF-C2 (future) runs
- We plan to assess the causes of differences between OH in the CCMs.
- Use of the box model enables us to distinguish between differences due to chemical mechanism and those due to differences in OH precursors.
- We can also use this output to predict future trends in OHTROP, based on CH4, H2O, and overhead O3 from the future CCM runs.