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

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1. Scientific motivation for studying tropospheric OH

Sensitivity of OH^{TROP} to climate change

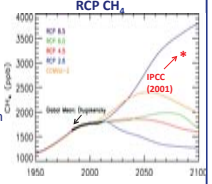
- IPCC (2001) predicted future OH would fall due to rising CH_4
- Best knowledge of global tropospheric OH (OH^{TROP}) comes from analysis of CH_3CCl_3 observations
- The CH_4 lifetime inferred from OH^{TROP} based on CH_3CCl_3 is 8.9 years; yet many sources use a CH_4 lifetime of ~12 years, which was found by modeling studies described in IPCC (2001)
- The 12 year CH_4 lifetime is central to subsequent IPCC reports and is called the "perturbation lifetime"
- Our work is motivated by understanding:
  - what factors other than rising CH_4 will affect OH^{TROP}
  - veracity of notion that future OH^{TROP} will decline

Importance of OH^{TROP}

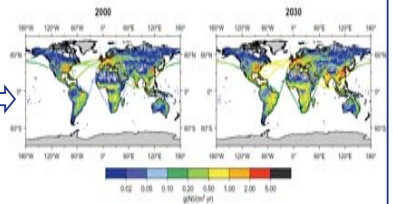
- OH serves as the main sink for many species: CO, CH_4, CH_3CCl_3, all HCFCs and HFCs, biogenic VOCs (isoprene and terpene), anthropogenic VOCs (formaldehyde, benzene, toluene, etc.)
- HO_x is integral to O_3 formation and destruction pathways; alone, HO_x depletes O_3, but with NO_x and VOCs, HO_x creates O_3

Methane Oxidation and OH

- Under low NO_x, the oxidation of CH_4 destroys HO_x
- Under high NO_x, the oxidation of CH_4 produces HO_x
- **Loss of methylperoxy radical CH_3O_2 with NO or HO_2 is critical**
- Future levels of CH_4 and NO_x are highly uncertain
  - CH_4 between now and 2100 varies dramatically among RCP scenarios
  - NO_x 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 OH^{TROP} will also depend on overhead column O_3, local humidity, and biogenic emission of VOCs, etc.
- ***Given this complexity, we expect CCMs to project a wide range of values for OH^{TROP} during the rest of this century***



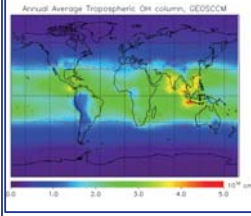
NO_x emissions from industry, power generation, traffic, domestic heating, and biomass burning used as input for prior CCM calculations, for years 2000 (38.0 Tg N / yr total) and 2030 (67.6 Tg N / yr total). From Eyring et al., *ACP*, 2007.

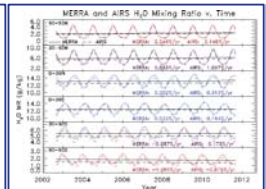


2. Methods for estimating changes in OH^{TROP}

Model OH

- Initial modern-day conditions for OH are taken from a time-slice run of GEOS CCM using 2005 emissions
- 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 H_2O, overhead O_3 and tropical widening are found from analysis of initial OH field



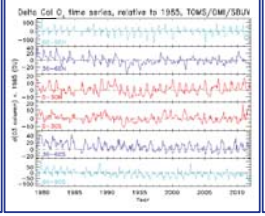


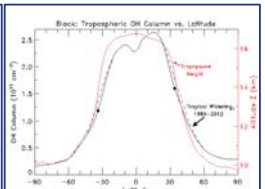
MERRA & AIRS H_2O

- Specific humidity files from the NASA Modern-Era Retrospective analysis for Research and Applications (MERRA) starting prior to 1985, &
- H_2O mixing ratio files from the NASA Atmospheric Infrared Sounder (AIRS) starting in 2002 were used to establish trends in H_2O by latitude
- OH^{TROP} is assumed to follow the square root of the change in H_2O using a steady-state assumption
- We plan to refine the effect of changing H_2O on OH^{TROP} by examining reaction rates from archived runs of GEOS CCM

Overhead O_3

- Total column O_3 trends were obtained from the NASA merged O_3 data set, consisting of measurements from SBUV, TOMS, and Aura OMI instruments
- We then use our photolysis code to estimate the impact on $J(O_3) \rightarrow O(^1D)$ of decreasing initial GEOS CCM overhead O_3 columns by amount suggested by the NASA product
- OH^{TROP} is assumed to change by the square root of $J(O_3) \rightarrow O(^1D)$





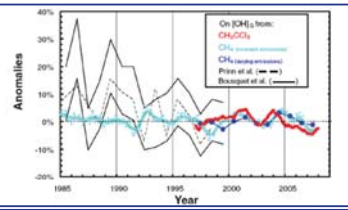
Tropical Widening

- Widespread evidence for expansion of the latitudinal circulation system (e.g. Seidel *et al.*, 2008)
- GCMs forced by GHGs cannot reproduce extent of tropical widening (Johanson and Fu, 2009)
- Allen et al. (2012) have suggested tropospheric O_3 and BC are responsible
- We simulate tropical expansion by increasing OH near the tropical boundary by a factor representative of 2° widening/decade (globally)

3. Preliminary results for expected changes in OH^{TROP}

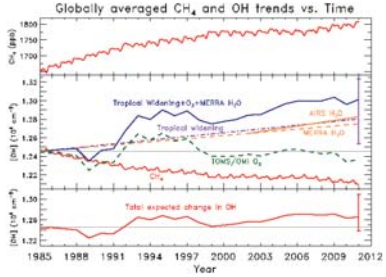
Previous Work on OH^{TROP} Trends

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



Current Conclusions from OH^{TROP} Analysis

- Effect of CH_4 on OH^{TROP} is taken from IPCC (2001), Section 4.2.1.1, which states that "the feed-back of CH_4 on tropospheric OH" found using contemporary chemical transport models is –0.32% for every 1% increase in CH_4 (red line, middle panel of figure to right)
- Primary effect of overhead O_3 is rise in OH^{TROP} following the 1991 eruption of Mount Pinatubo (green dashed line) due to enhanced removal of stratospheric O_3 by volcanic aerosol
- Rising H_2O from MERRA and AIRS increases OH^{TROP} (orange lines) by an amount comparable to the decrease expected from rising CH_4
- Widening of the tropics contributes an increase in OH^{TROP} (purple line) roughly equal to that due to rising H_2O, leading us to view this as a significant factor controlling OH^{TROP}
- Overall expected change in OH^{TROP} (bottom panel) shows higher level of interannual variability prior to ~1999 and lower variability thereafter



4. Future plans for refining estimates of ΔOH^{TROP}

Improve Estimate of $d(OH^{TROP}) / d(H_2O)$

- Reaction rates from recent runs of GEOS CCM are archived for reactions such as:

  $H_2O + O(^1D) \rightarrow 2OH$

- Using these reaction rates we will determine the proportion of OH that is produced via reaction with H_2O
- The determined scaling factor would be used to calculate a new ΔOH^{TROP} based on the H_2O trends
- Estimate time- and pressure-varying values of $d(OH^{TROP}) / d(H_2O)$
- Evaluate discrepancies between MERRA and AIRS H_2O trends

Evaluate CH_4/OH Feedback

- We will use a box model (details below) to probe relationship between CH_4 and OH^{TROP} and its dependence on NO_x

Box Model

We will use the Dynamically Simple Model of Atmospheric Chemical Complexity (DSMACC) box model (provided by Mathew Evans) with chemical mechanistic information taken from the Master Chemical Mechanism, MCM v3.2 (Jenkin *et al.*, 1997; Saunders *et al.*, 2003) via website http://mcm.leeds.ac.uk/MCM. Using the MCM, we have the capability to consider:

- the degradation of up to 135 VOCs
- the influence of $J(O^1D)$ and $J(NO_2)$ on OH chemistry
- the impact of NO_x on methane oxidation

Assessing OH^{TROP} 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-C1 (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 OH differences due to ***chemical mechanism*** and those due to ***differences in OH precursors***
- We can also use this output to predict future trends in OH^{TROP}, based on CH_4, H_2O, and overhead O_3 from the future CCM runs

Propagate Uncertainties

- Calculate uncertainties in AIRS and MERRA H_2O and NASA O_3 product
- Evaluate standard deviation in average fraction of OH production occurring via $H_2O + O(^1D)$
- Estimate uncertainty in the box model evaluation of $d(OH^{TROP}) / d(CH_4)$