Evaluating Effects of H₂O and Overhead O₃ on Global Mean Tropospheric OH Concentration

Julie M. Nicely¹, R. J. Salawitch¹, T. Canty¹, C. Lang², B. N. Duncan³, Q. Liang³, L. D. Oman³, R. S. Stolarski², D. W. Waugh²

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

1. Scientific motivation for studying tropospheric OH

Sensitivity of OH{TROP} to climate change

- IPCC (2001) predicted future OH would fall due to rising CH₃
- Better knowledge of global tropospheric OH (OH{TROP}) comes from analysis of CH₃CCl₃ observations
- The CH₃ lifetime inferred from OH{TROP} based on CH₃CCl₃ is 8.9 years; yet many sources use a CH₃ lifetime of ~12 years, which was found by modeling studies described in IPCC (2001)
- The 12 year CH₃ lifetime is central to subsequent IPCC reports and is called the "perturbation lifetime"

Our work is motivated by understanding:
- what factors other than rising CH₃ will affect OH{TROP}
- the variance of notion that future OH{TROP} will decline

2. Methods for estimating changes in OH{TROP}

Base Case OH Conditions

- 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
- Calculated changes in OH due to H₂O and overhead O₃ are applied to initial OH field

MERRA & AIRS H₂O

• Specific humidity flux from the NASA Modern-Era Retrospective analysis for Research and Applications (MERRA) starting prior to 1985, and
• H₂O mixing ratio files from the NASA Atmospheric Infrared Sounder (AIRS) starting in 2002 were used to establish trends in H₂O by latitude
• OH{TROP} is assumed to follow the square root of change in H₂O using a steady-state assumption

Propagate Uncertainties

- Calculate uncertainties in AIRS and MERRA H₂O and NASA O₃ product

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

Overhead O₃ Observations

- Total column O₃ trends were obtained from the NASA merged O₃ data set, consisting of measurements from SBUV, TOMS, and Aura DMI instruments
- We then use our photolysis code to estimate the impact on (OH) / (O¹D) of decreasing initial GEOS CCM overhead O₃ columns by amount suggested by the NASA product
- OH{TROP} is assumed to change by the square root of (O¹D) / (O¹')

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

Improve Estimate of d(OH{TROP}) / d(H₂O)

- Reaction rates from recent runs of GEOS CCM are archived for reactions such as: H₂O + O¹D) → 2OH
- Using these reaction rates we will determine the proportion of OH that is produced via reaction with H₂O
- The determined scaling factor would be used to calculate a new ΔOH{TROP} based on the H₂O trends
- Estimate time- and pressure-varying values of d(OH{TROP}) / d(H₂O)
- Evaluate discrepancies between MERRA and AIRS H₂O trends

Evaluate CH₄/OH Feedback

- We will use a box model (details below) to probe the relationship between CH₄ and OH{TROP} and its dependence on NOₓ

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 (hindsight) 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 CH₄, H₂O, and overhead O₃ from the future CCM runs

Box Model

We will use the GSCF Combined Stratosphere-Troposphere (COMBOS) box model provided by Chang Lang (JHU):
- GMI chemical mechanism
- 118 species, 321 thermal reactions, and 81 photolysis reactions
- 5 modules:
  - Aerosol optical depth & surface area
  - Photolysis scheme
  - Thermal reactions scheme
  - Differential quantum yield
  - Input-output
- Fast-J photolysis & MIVGAR II solver

Evaluate standard deviation in average fraction of OH production occurring via H₂O + O¹D)
- Estimate uncertainty in the box model evaluation of d(OH{TROP}) / d(CH₄)

Current Conclusions from OH{TROP} Analysis

- Effect of CH₄ on OH{TROP} 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, middle panel of figure to right)"
- Primary effect of overhead O₃ is rise in OH{TROP} 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 OH{TROP} (orange lines) by an amount comparable to the decrease expected from rising CH₄
- Overall expected change in OH{TROP} (bottom panel) shows higher level of interannual variability prior to ~1999 and lower variability thereafter

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 Bouquet 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